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ABSTRACT

Analytical and experimental studies in 1958 through 1960 investigated the instrumentation required for detecting the radioactive wakes of submarines. Volume distributed spectra for sodium-24, chlorine-38, and potassium-40 were measured for various sized sodium iodide crystals and plastic phosphor gamma scintillation detectors. Chemical methods of concentrating the radioactive samples, and of selectively discriminating against the background radioactivity indicated vaporization and ion-exchange techniques as the more practical.

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SUMMARY AND CONCLUSIONS

There are at least two primary unknowns which need to be resolved before the question of feasibility can be decided:

- a. What are the energy and the radial distribution of neutron leakage from the submarine's hull? (C)
- b. What are the dynamic characteristics of the submarine's wake?

The volume distributed spectra for sodium-24, chlorine-38, and potassium-40, were examined under laboratory conditions. Spectral data are presented for the following scintillator materials and dimensions: (U)

Sodium Iodide (thallium activated) 2 x 1 $\frac{1}{2}$, 3 x 3, and 5 x 4 inches

Plastic Phosphor Pilot B 3 x 3 and 5 x 4 inches

The data indicates that energy discrimination techniques are likely to improve the chances for success in counting experiments. The degree of improvement depends upon the concentration of the induced radionuclides to the major background contributor, the natural potassium-40 content of the ocean. An attempt was made to predict the effect of large size detectors on signal but additional experimental work is required to formulate the engineering requirements. (U)

Section 3 is devoted to the various means of chemically concentrating the signal source or depressing the background. Six chemical concentration techniques are described: vaporization, ion-exchange, precipitation, freezeout techniques, liquid-liquid extraction, and membrane processes. Of these, vaporization and ion-exchange techniques show some potential. (U)

Vaporization techniques remove all or part of the water from sea water samples.

Two schemes were considered in detail:

- a. Reduction of a sea water sample to a saturated solution (liquid phase detection). (U)
- b. Reduction of sea water sample to a dry solid (solid phase detection). (U)

The computed concentration ratios attainable for "a" above, are 7.4 on a weight basis and 9.75 on a volume basis, and for "b" above, 28.5 on a weight basis and 59.1 on a volume basis. (U)

Ion-exchange techniques can be used to reinforce nuclear detection by either of the following methods: (U)

- a. Reduction of sea water sample volume by taking the desired ion from sea water and retaining the ion in a more concentrated state or releasing the ion in a more concentrated solution. (U)
- b. Reduction of the natural background radioactivity by removing all or part of the radioactive background. (U)

The computed concentration ratios (on a volume basis) attainable by detailed ion-exchange methods are:

- a. Concentrate on the resin and detect on resin = 2. (U)
- b. Concentrate on resin and detect in regenerating solution = 5. (U)
- c. Concentrate activity in the regenerating solution, evaporate to dryness and detect = 66. (U)

The essence of these methods is to reduce the volume -- hence increasing the specific activity with the concomitant increase in counting rate for a given volume and a reduction in background counting rate resulting in considerable decrease in counting time required for a given statistic. (U)

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System requirements will have to be determined once the operational objectives are called out. It is not suggested that chemical analysis be carried out on an ocean search problem but rather as a means of strengthening the search systems potency. (C)

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INTRODUCTION

The wide use and acceptance of the scintillation method for examining complex gamma ray spectra suggested that this technique be employed in the submarine detection problem. The activation of sea water by thermal neutrons results in the production of at least two radioisotopes, sodium-24 and chlorine-38, which have decay schemes which could lend themselves as a signal source. The technological progress that has been made during the last ten years in growing large single crystals of the alkali halides together with the plastic and liquid scintillator materials has focused attention on the use of these materials for threshold detection purposes. Further, the development of end-window photomultiplier tubes with low noise characteristics and uniform photocathodes of relatively large surface area together with improvements in pulse amplifiers and pulse height analyzers were necessary complements to the scintillation detection methods. (C) (U)

Section 2 describes the theoretical considerations of induced radioactivity in sea water and experimental results. Discussion on detection techniques is limited to the nuclear interactions of the escaping neutrons from a nuclear submarine hull with the surrounding sea water. (C)

No consideration is given at this time to other nuclear interaction mechanisms such as: neutron detection, cosmic ray anomalous effects, Cherenkov radiation, molecular beam methods, microwave absorption spectroscopy, nuclear magnetic resonance, optical spectroscopy, and mass spectroscopy. (C)

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Related experimental work is being carried out by the Naval Air Development Center at Johnsville, Pennsylvania, and by the Hudson Laboratories in New York. The work reported in this Section supplements their work by providing basic data on volume distributed sources for different scintillating materials with a variety of dimensions. (U)

Section 3 describes the concentration and detection of induced radioactivity in sea water. It is believed that radioactivity artificially induced in sea water may be concentrated by chemical means to a level which will permit detection with apparatus now available. The methods examined reduce detection techniques to two bases: (U)

- a. Concentration of the activity present in a sea water sample into a relatively small volume to improve counting geometry. (U)
- b. Removal of the radioactive background in a sea water sample in order to improve the counting statistics. (U)

Six chemical processing techniques will be discussed. These are: Vaporization, Ion-Exchange, Precipitation, Freezing, Liquid-Liquid Extraction, and Membrane Processes. (U)

This report was submitted in compliance with contract Nonr-2777(00), Project NR 220-012 of the Office of Naval Research and summarizes the work completed during the period January 1, 1959 through June 30, 1960. (U)

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IN SEA WATER AND EXPERIMENTAL RESULTS

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1. OBJECTIVE

The objective of this Section is to consider the practicability of detecting radioactivity induced in the wake of a nuclear powered submarine with a sea-going gamma-sensitive system. The interaction of neutrons escaping through the submarine hull with the surrounding sea water is to be investigated. (C)

Detailed objectives are to:

1. Calculate the activity resulting from flash activation of sea water by various levels of neutron leakage through a submarine hull. (C)
2. Provide data from which estimates can be made of the gamma-ray energy spectrum of the activated sea water and its change with time. (U)
3. Provide data on the response of various scintillation detectors to volume - distributed sources of potassium-40 which is the principal gamma emitter in sea water, and of chlorine-38 and sodium-24 which would be produced by neutron activation of sea water. On the basis of this information, a final selection can be made of detector size and composition. (U)

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2. THE CHEMISTRY OF SEA WATER (Ref. 1)

Table 1 is a partial list of the elements present in sea water. The data is reported for a chlorinity of 19⁰/oo. The empirical relationship between salinity and chlorinity was established by an Internal Commission as:

$$\text{Salinity} = 0.03 + 1.805 \times \text{chlorinity}$$

Chlorinity is a defined quantity which closely approximates chloride content of sea water. Both chlorinity and salinity are always expressed in terms of grams per kilogram of sea water; i.e., in parts per thousand or per mille (analogous to per cent), and the symbol is ⁰/oo.

The relative proportions of the different major constituents are virtually constant in the open ocean and are independent of the absolute concentration. The exceptions occur in regions of abnormal salinity (high dilution or evaporation). This is an important consideration in that the relative amounts of sodium-24 and chlorine-38 initially produced should remain constant as these are the principle neutron activation products. The minor constituents are more subject to variations caused by dilution, evaporation, and various biological, chemical, and geological processes.

Some pertinent definitions are:

Chlorinity: Originally defined as the total amount of chlorine, bromine, and iodine in grams contained in one kilogram of sea water, assuming that the bromine and iodine had been replaced by chlorine. More recently the chlorinity has been defined as identical with the number of grams of "atomic weight" silver needed to precipitate the halogens in 0.32285235 kilograms of the sea water sample. This latter definition is independent of revisions in the accepted values of the atomic weights.

TABLE 1. Partial Table of Elements Present in Sea Water (Ref. 2)
(Chlorinity = 19.00 ‰)

<u>Element</u>	<u>mg/kg (Weight of Element per Kilogram of Sea Water)</u>
Chlorine	18980
Sodium	10561
Magnesium	1272
Sulphur	884
Calcium	400
Potassium	380
Bromine	65
Carbon	28
Strontium	13
Boron	4.6
Fluorine	1.4

Salinity: The total amount of solid material in grams contained in one kilogram of sea water when all the carbonate has been converted to oxide, the bromine and iodine replaced by chlorine and all organic matter completely oxidized.

In addition to Table 1 which is a partial listing of elements present in sea water, the following values are used in calculations in this report:

Chlorine - 37

$$\begin{aligned}\rho &= 0.54803 \text{ gm-atoms/liter} \\ a_f &= 0.246 \\ \sigma_a &= 0.56 \text{ barns} \\ \lambda &= 3.1 \times 10^{-4} \text{ per sec.} \\ \Lambda &= 36.977 \text{ gms}\end{aligned}$$

Sodium-24

$$\begin{aligned}\rho^* &= 0.45917 \text{ gm-atoms/liter} \\ a_f &= 1.0 \\ \sigma_a &= 0.536 \text{ barns} \\ \lambda &= 1.282 \times 10^{-5} \text{ per sec.} \\ \Lambda &= 22.996 \text{ gms}\end{aligned}$$

* Total chlorine and total sodium content for sea water of 19.00 chlorinity. (Ref. 1)

3. THE SEA AS A NUCLEAR ENVIRONMENT

3.1 BACKGROUND ACTIVITY

An important factor in considering any low level counting problem is the background contribution to the signal. There are a number of sources of background for the problem under consideration which symbolically can be represented as:

$$B = B_1 + B_2 + \dots + B_n$$

B_1 = potassium-40 and other naturally occurring radioactive nuclides in sea water

B_2 = potassium-40 in sodium iodide crystal

B_3 = potassium-40 in glass envelope of photomultiplier and other contaminants in structural material

B_4 = cosmic ray

B_5 = fallout

B_6 = concentration of radio-nuclides in marine life

B_7 = instrument noise

B_8 = shipboard contamination and other sources of contamination

In this section we will consider some of these background sources and attempt to evaluate their relative importance.

3.2 NATURAL RADIOACTIVITY

3.2.1 SEA WATER

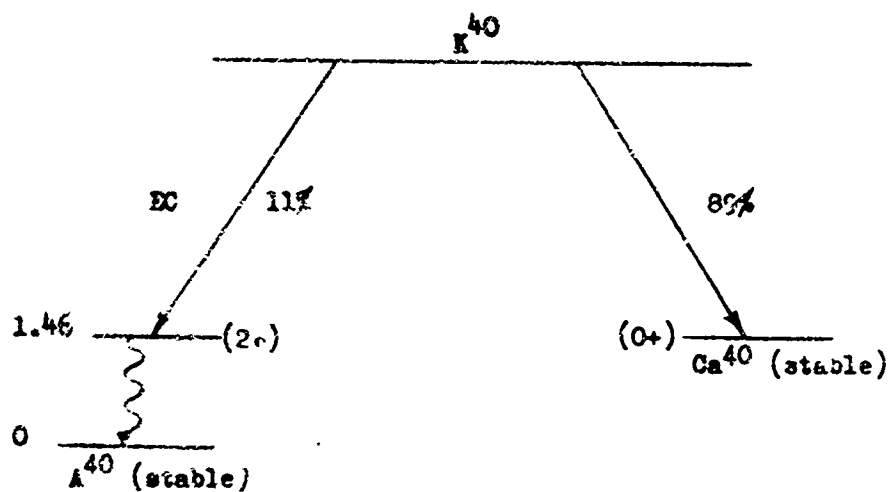
Table 2 exhibits the naturally occurring radio-nuclides in sea water together with concentration, specific activity, and total ocean content with respect to weight and activity. The potassium-40 content seems to be the principal source of background with a specific activity of 1.2×10^{-2} disintegrations per sec. per ml. Figure 1 shows the decay scheme for potassium-40 (Ref. 3). The associated gamma

TABLE 2. Radioactivity of Sea Water (Ref. 4)

Radionuclide	Concentration $\mu\text{Ci/cm}^3$	Specific Activity (Disintegrations/sec/cm ³)	Total Amount in Ocean Megatons	Total Activity in Ocean Megacuries
⁴⁰ K	4.5×10^{-8}	1.2×10^{-2}	63,000	460,000
⁸⁷ Rb	8.4×10^{-8}	2.2×10^{-4}	118,000	8,400
²³⁸ U	2.0×10^{-9}	1×10^{-4} (a)	2,800	3,800
²³⁵ U	1.5×10^{-11}	3×10^{-6} (a)	21	110
²³² Th	1×10^{-11}	2×10^{-7} (a)	14	8
²²⁶ Ra	3.0×10^{-16}	3×10^{-5} (a)	4.2×10^{-4}	1,100
¹⁴ C	4×10^{-17}	7×10^{-6}	5.6×10^{-5}	270
³ H (b)	8×10^{-20}	2.5×10^{-5}	1.5×10^{-3}	12
¹³⁸ La	2.5×10^{-13}	7.4×10^{-11}	0.35	2.6×10^{-3}

Note (a) activity of nuclide + daughter products.

Note (b) only in top 50-100 meters of the ocean.



$$Q_{EC} = 1.33 \text{ Mev}$$

$$\gamma = 1.46 \text{ Mev}$$

$$T_{1/2} = 1.3 \times 10^9 \text{ years}$$

FIGURE 1. Decay Scheme of Potassium-40 (Reference 3)

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activity (1.46 Mev energy) for this isotope is approximately 3.27×10^{-5} microcuries per milliliter. The values quoted are typical and it remains to be determined what the variation from one geographical location to another may be, also, what variation exists with respect to depth for any given location. The closest competitor of potassium-40 is rubidium-87 which is a factor of approximately 50 below the potassium-40 specific activity. This isotope is a pure beta emitter and is not expected to contribute a background signal. Thorium C" (Th^{208} , 2.62 Mev., daughter in the thorium series) may be significant for extremely low signal levels. The potassium-40 integral counting rates for sodium iodide (thallium activated) crystals of various sizes and the differential spectra are discussed in Section 3.3.1 of this report. (U)

3.2.2 COSMIC RAYS

A quantitative discussion of cosmic ray background must be deferred pending proposed measurement programs. However, some estimate of the relative activity as a function of depth can be obtained from Figure 2. Figure 3 shows the recorded cosmic ray background for a 4×1.5 inch NaI(Tl) crystal under 8 inches of steel + 2 inches of lead + 1 inch of triple distilled mercury (Ref. 5). On a volume basis, extrapolation to a 5×4 inch NaI(Tl) crystal with an equivalent water thickness the integral counting rate (above 100 kev) is estimated to be approximately 43 cps while integral counting rate above 1.5 Mev should yield 3 cps (Figure 4). This is a hazardous extrapolation at best and the statements made above should be treated with this in mind. (Later field measurements have shown this estimate to be much too low, - see Boeing Document D2-7474 Field Test on Nuclear Submarine Detection - August 1960). (C)

It is not possible to eliminate the cosmic ray component entirely with a reasonable thickness of material because of the extremely

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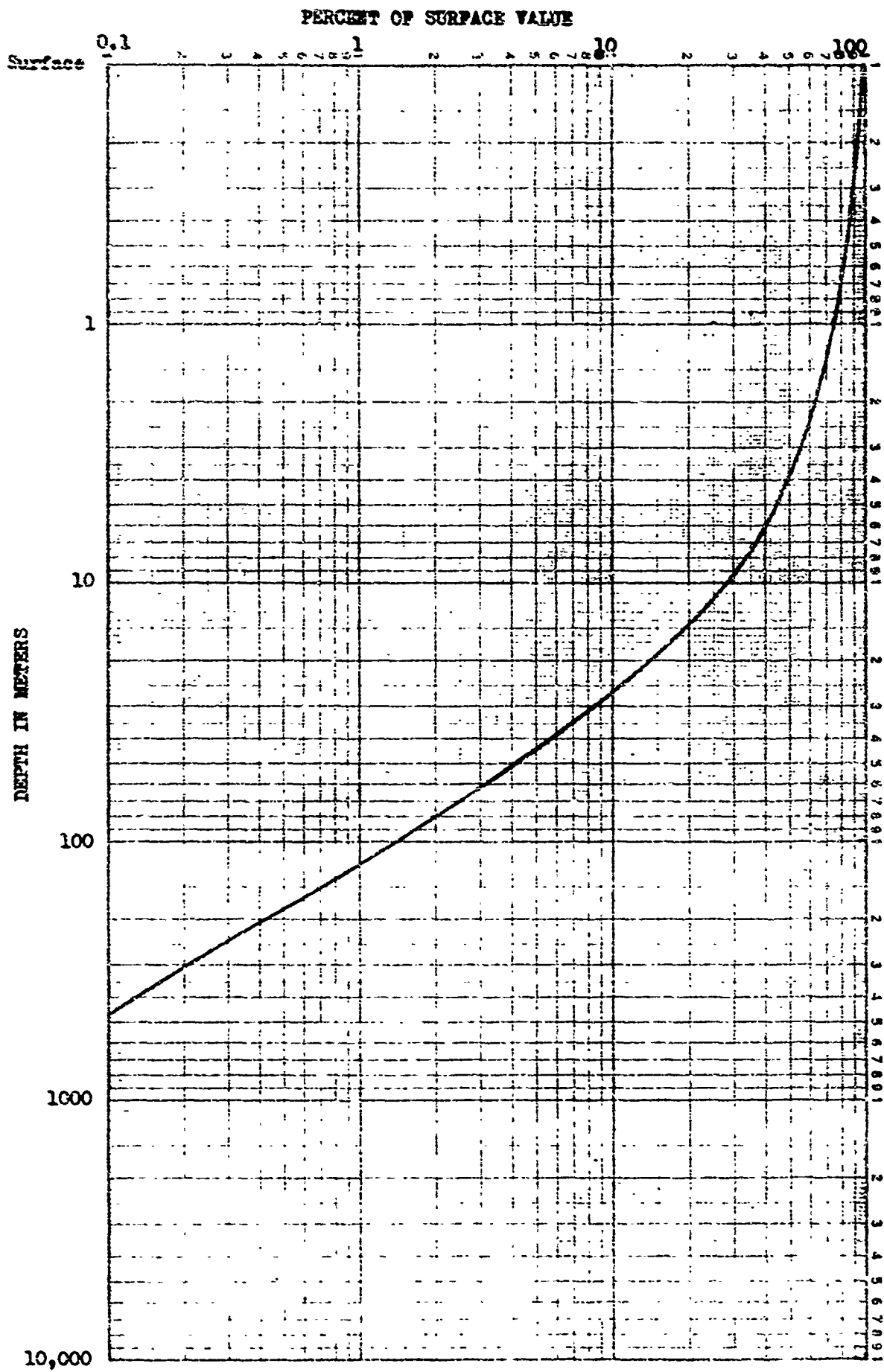
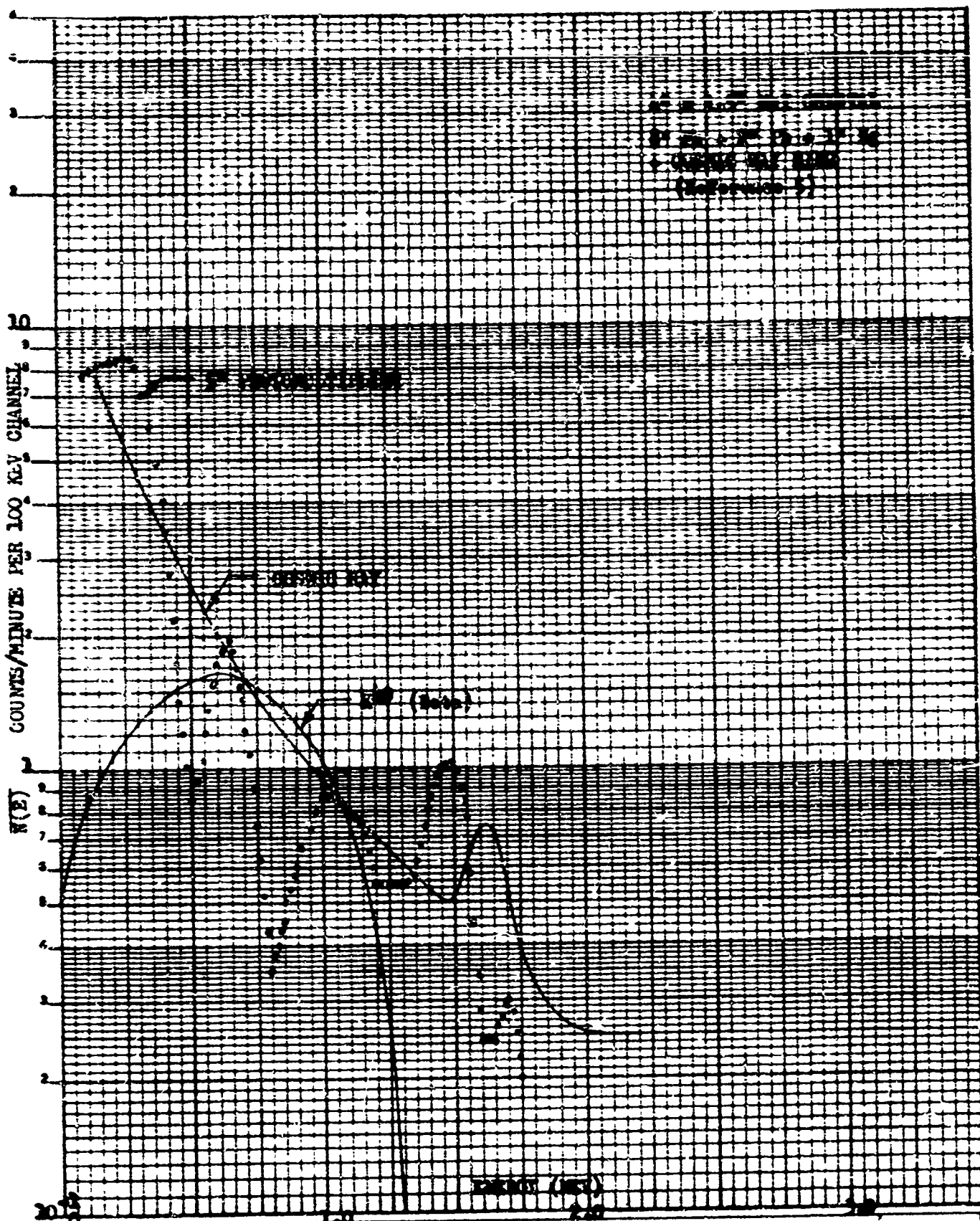
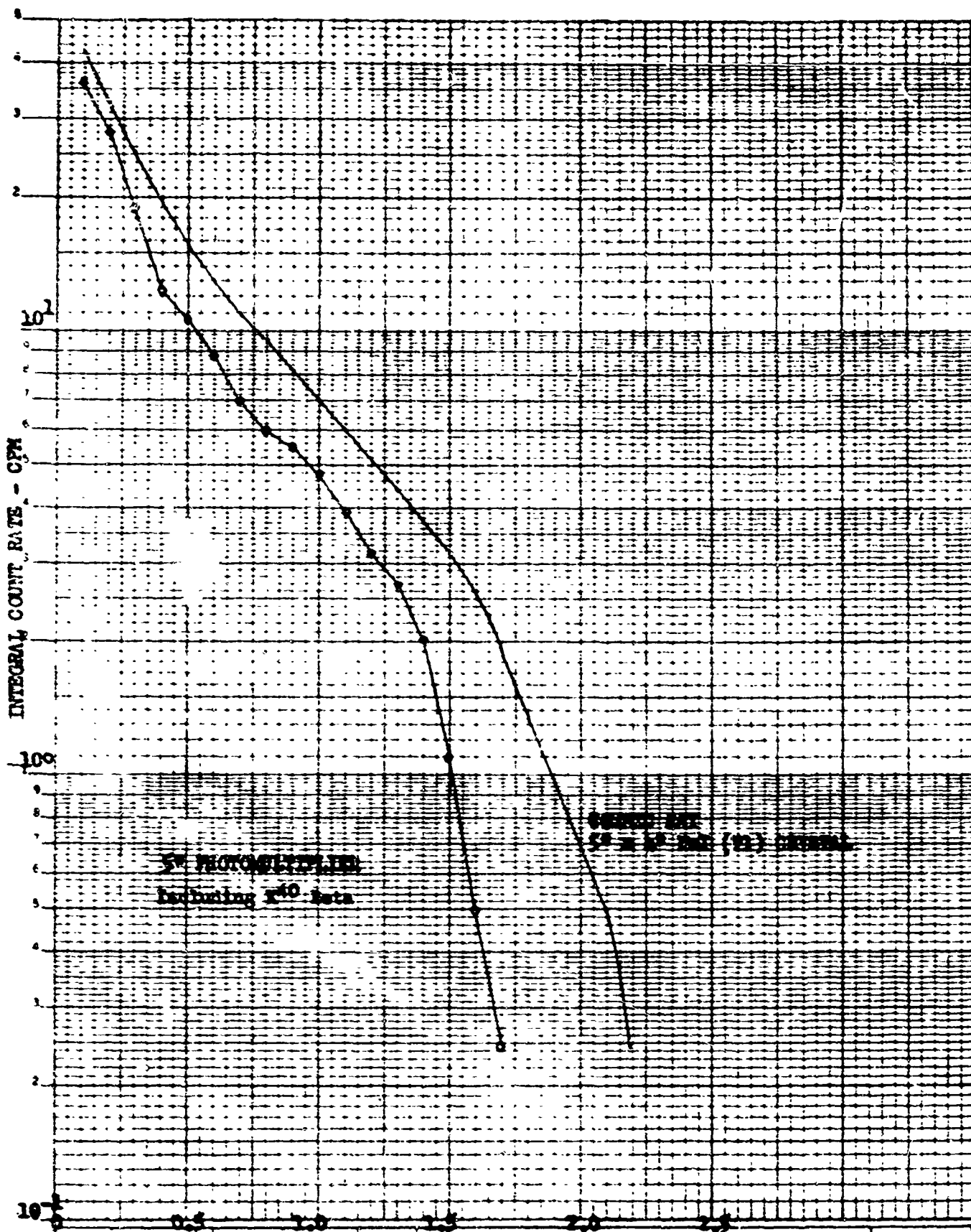


Figure 2. Attenuation of Cosmic Rays in the Sea (Ref.24)



CALC		REVISED	DATE	Figure 3. Differential Background Spectra due to K^{40} beta particles, cosmic rays, and the 5" photomultiplier gamma rays. BOEING AIRPLANE COMPANY	D2-7908
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CALC CHECK APR APR	REVISED 	DATE 	ENERGY - Mev Figure 4. Integral Background Spectra Summed and Extrapolated from Figure 3. BOEING AIRPLANE COMPANY SEATTLE 24, WASHINGTON	D2-7908 Sec. 2. PAGE 17 OF
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high energy of the cosmic ray background. If the background contribution from this source is objectionable it could perhaps be eliminated with the use of a cosmic ray guard umbrella. This recourse would result in added complexity of electronic equipment. The umbrella could consist of a number of cosmic ray sensitive detectors and the count resulting from the incident particle would be electrically subtracted from the total count by an anticoincidence circuit. That is, any cosmic particle which would result in a signal from the sample detector circuit would by virtue of the geometrical configuration also have to pass through the guard circuit. The resultant signal is simultaneous to both detectors and would be electrically annulled.

3.3 INSTRUMENTATION PROBLEMS

3.3.1 CRYSTAL

Potassium-40 is a known contributor to the intrinsic crystal background and is found in concentrations of 3 ppm. The crystal growing industry is cognizant of this fact and some control can be exercised by careful crystal selection. An unknown source of crystal background can be traced to crystal contamination and is thought to be cesium-137 (fission product) (Ref. 6). This source is estimated to contribute 210 counts per minute for a 5 x 4 inch NaI(Tl) crystal, but should be of no consequence for integral counting above 0.66 Mev. The use of stainless steel No. 304 as a canning material as well as the use of quartz between photomultiplier and crystal results in some reduction of background. Stainless steel No. 304 is reported to possess an alpha background of $0.019 \pm .012$ counts per minute per 20 square centimeters (Ref. 7); quartz is reported to possess an alpha background of $0.003 \pm .007$ counts per minute per 20 square centimeters (Ref. 8).

3.3.2 PHOTOMULTIPLIER

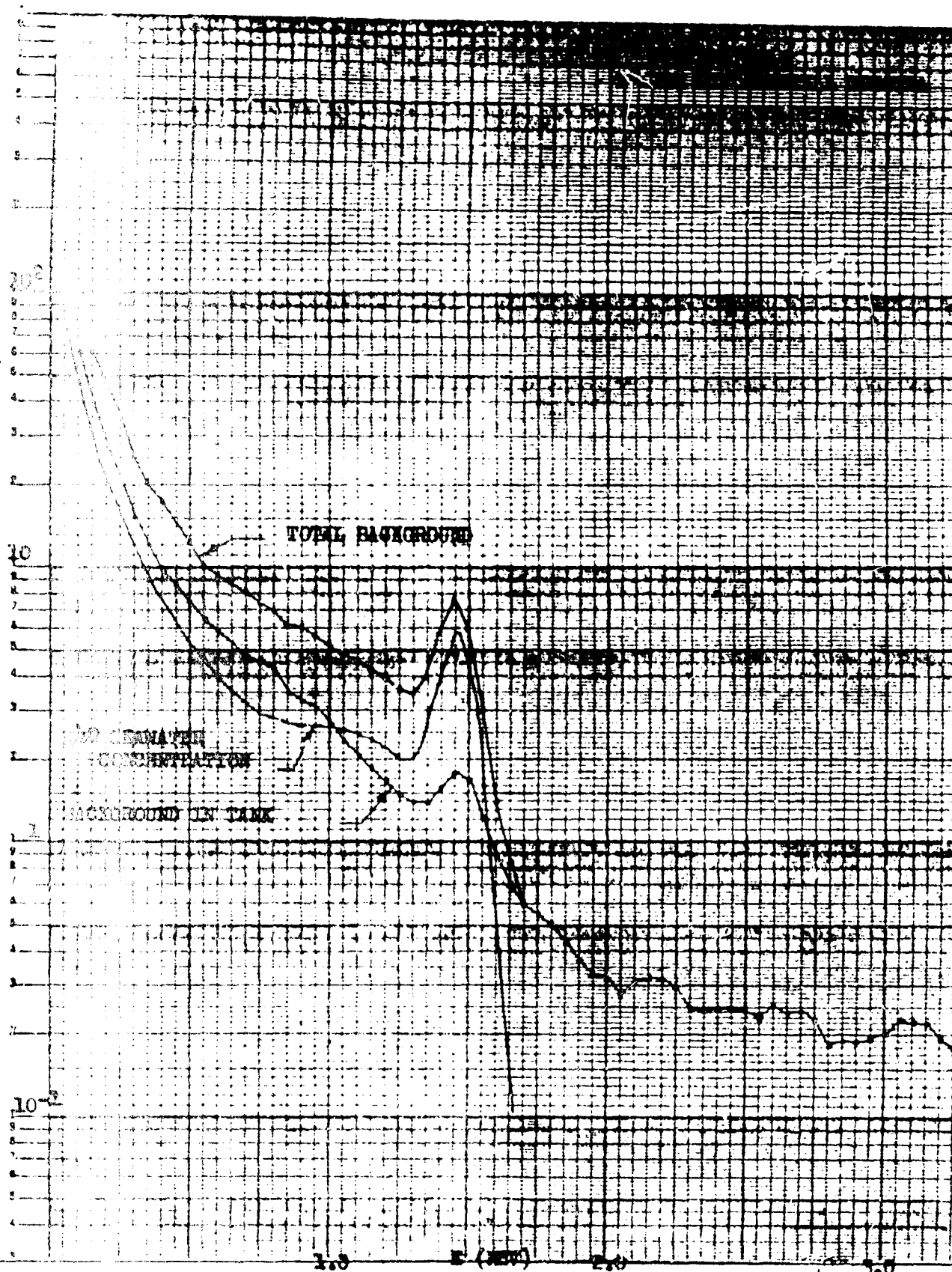
Contaminents are present in the photomultiplier itself. This source of background originates in the glass envelope of the phototube and in the tube base material. The radio elements are potassium-40 and radium. No activity has been reported emanating from the internal tube structure.

The activity of the photomultiplier varies directly as the mass. Tube manufacturers state that natural potassium, which emits 3 gammas per second per gram, constitutes about a tenth of one percent of the weight of the glass of the phototube. The degree of contamination will depend on the sample's past processing history. Reports indicate that aluminum samples assay at approximately 3×10^{-13} curies of radium per gram of metal (Ref. 9). This appears to be true and is independent of the vendor and applies to 28 as well as 17 ST, 52 ST and 61 ST, alloy stocks. Iron is relatively clean when compared with lead. The smelting and processing procedures eliminate most of the radio-activity as oxides which are expelled with the slag material. Some plastics are generally free from radioactive contaminants but their feasibility for use in seagoing probes remains to be evaluated. With respect to finishing materials leaded paints should be avoided because of the possibility of lead-210 (Radium D) contamination.

3.4 BACKGROUND MEASUREMENTS

Background measurements were made with the 5 x 4 inch NaI(Tl) probe in the 2500 gallon tank filled with water (Part 6). The differential spectrum which was obtained is shown in Figure 5 together with the spectrum of a volume distributed source of potassium-40 at a concentration

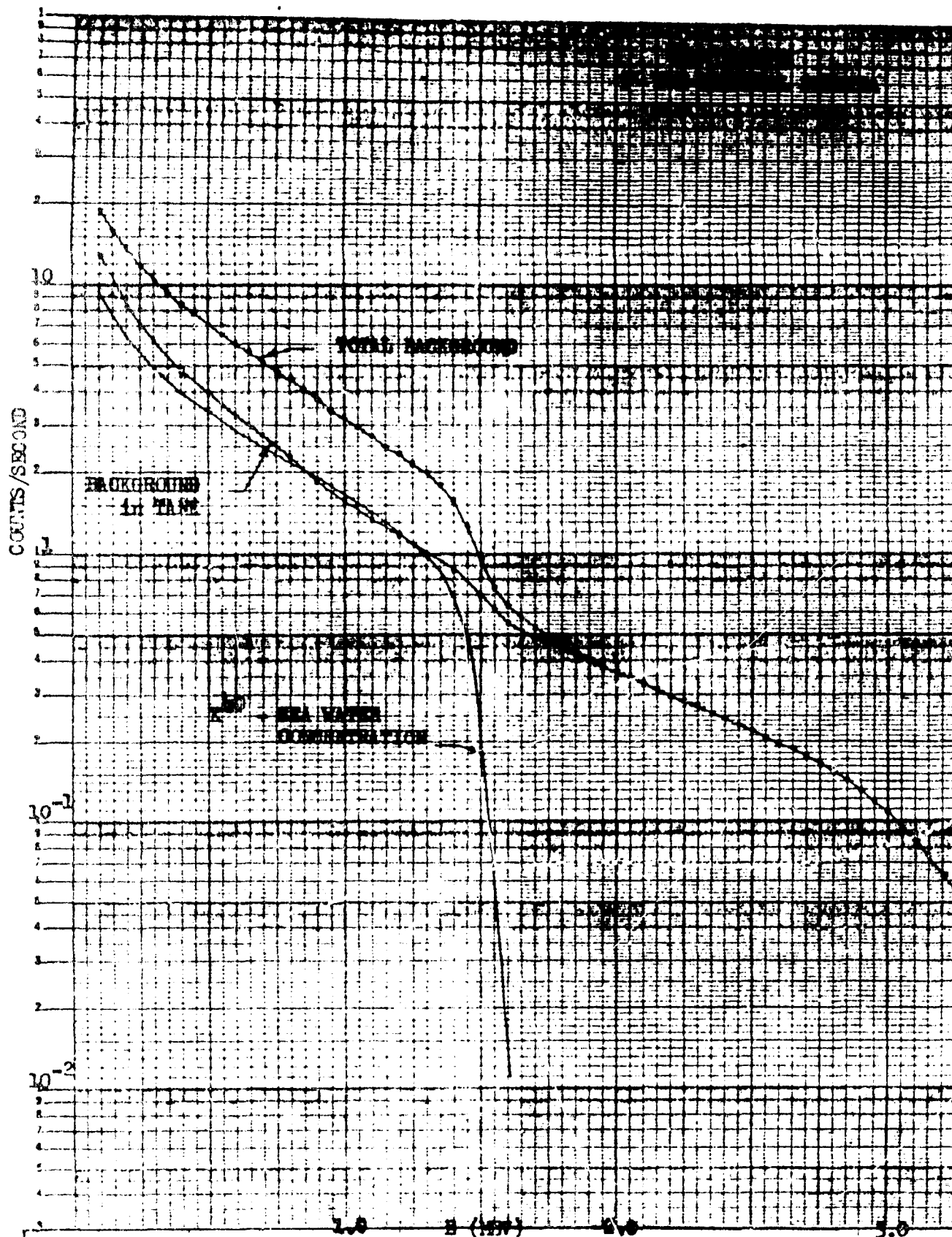
NET COUNTS/SECOND/KEV



REVISED	DATE	Figure 5. Differential Background Spectra in the Tank	D2-7908
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corresponding to that of sea water. These curves were added and the sum curve is also plotted in Figure 5. The corresponding integral curves are shown in Figure 6. Insofar as the room background approximates the background of the sea, these are the spectra which would be obtained with this probe in the ocean. It is reasonable to expect that these represent an upper limit to the background spectrum which would be obtained at sea. The intrinsic background of the probe can be reduced as explained above; the contribution from the radioactivity present in the concrete walls of the counting room would not be present; and except for the surface region the cosmic ray contribution would be less and a function of distance below the sea surface.



DESIGNED DRAWN APPR ADM	REVISED DATE	Figure 6. Integral Background Spectra In the Tank	D2-7908
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4. THERMAL NEUTRON INDUCED RADIOACTIVITY IN SEA WATER

4.1 FLASH ACTIVATION

In this section we will consider the results of a flash (instantaneous) activation of one liter of sea water by one gram-atom of thermal neutrons. The problem is approached in this manner so as not to introduce any submarine characteristics such as speed and active hull length, but rather to obtain some estimate of the relative magnitudes of the reaction products. All of the calculations have been carried out assuming a value of chlorinity equal to 19 ‰ with a resulting salinity of approximately 34.5 ‰. These values are believed to be representative of the problem. (C)

The calculations which follow are similar to those performed in the paper by D. Dryssen and P. C. Nyman (Ref. 10). The above mentioned paper, however, did use slightly different values for the concentrations of various elements which, together with some typographical errors and omissions of some known radioisotopes, justifies a re-examination of the results. (U)

The number of nuclei, N , of a particular radioisotope as a result of the absorption of B thermal neutrons is given by: (U)

$$N = \frac{\sum_{j=1}^Z C_j' \cdot (\sigma_{act})_j \cdot B}{\sum_{i=1}^Z C_i \cdot (\sigma_{ab})_i} \quad \text{gram-atoms}$$

where:

C' is the number of gram-atoms per liter of the isotope which produces the active isotope.

C is the number of gram-atoms per liter of the element whose isotopes absorb neutrons.

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σ_{act} is the thermal neutron activation cross section, expressed in units of barns.

σ_{abs} is the thermal neutron absorption cross section, expressed in units of barns.

The summation index, $j=1$ to q , accounts for the number of ways in which the active isotope can be formed, i.e., (n,p) , (n,α) , (n,γ) .

The summation index, $i=1$ to n , accounts for the total number of elements undergoing neutron absorption reactions.

The product $N\lambda$ then gives the disintegration rate (disintegrations per second) of the active isotope. Dividing $N\lambda$ by 3.7×10^{10} converts the disintegration rate to the number of curies of the radioisotope in question. The activity, A , in curies at time $T=0$ is:

$$A(0) = \frac{N\lambda}{3.7 \times 10^{10}} = \frac{\sum_{j=1}^q C_j' (\sigma_{act})_j}{\sum_{i=1}^n C_i (\sigma_{abs})_i} \times \frac{\lambda}{3.7 \times 10^{10}} \text{ curies}$$

where λ is the decay constant, expressed in units per second.

To find the activity in curies for the isotope of interest as a function of time ($T \geq 0$), the following relation is used:

$$A(T) = A(0) e^{-\lambda T} \text{ curies}$$

where T is the time elapsed after irradiation takes place, expressed in units of seconds.

The activity at $T=0$ is high for some isotopes (see Table 3). It can be seen that those isotopes with a particularly short half life

(lithium-8, boron-12, fluorine-20, oxygen-19, aluminum-28) contribute an insignificant amount to the total activity after the first tenth of an hour.

Figure 7 depicts the total activity as well as the individual activities as a function of time after irradiation ceases for those radioisotopes whose activity at T=0 is one curie or more. It can be seen that chlorine-38 is the largest single contributor during the first 1.8 hours. Thereafter sodium-24 is predominant for the next 99 hours, after which the total activity is due to sulfur-35.

Figure 8 shows the activity for the short half-life isotopes lithium-8, boron-12, fluorine-20, and oxygen-19. These were not shown in Figure 3 so as not to detract from those isotopes which are of immediate interest for submarine wake detection. ($T_{1/2} \geq 1$ min.) and because they are of significantly lower activity.

It may be that those isotopes whose half lives are less than several minutes might have application to other than long range submarine detection such as type identification or tracking.

As noted previously the data shown in Figures 7 and 8 were made for a value of salinity equal to 34.5 ‰. The equations for salinities other than 34.5 ‰ will now be presented.

Since C and C' are proportional to the salt content, S, the following relations are valid within the limits discussed in part 2:

$$C_s = C_{34.5} \cdot \frac{S}{34.5}$$

$$C'_s = C'_{34.5} \cdot \frac{S}{34.5}$$

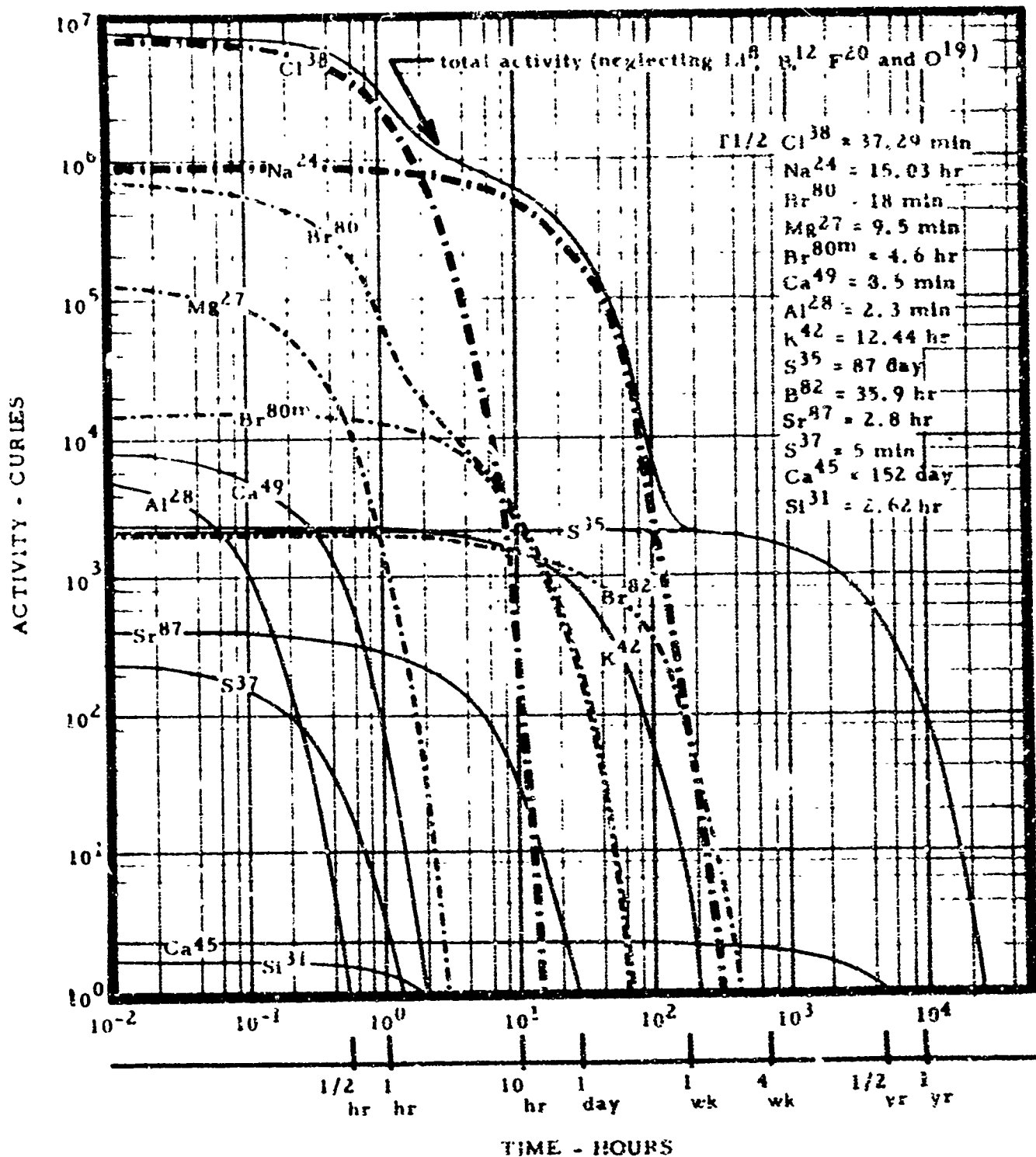


Fig 7 INDUCED ACTIVITY VS TIME
(Half Lives Greater than 2.3 Minutes)

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ACTIVITY - Curies

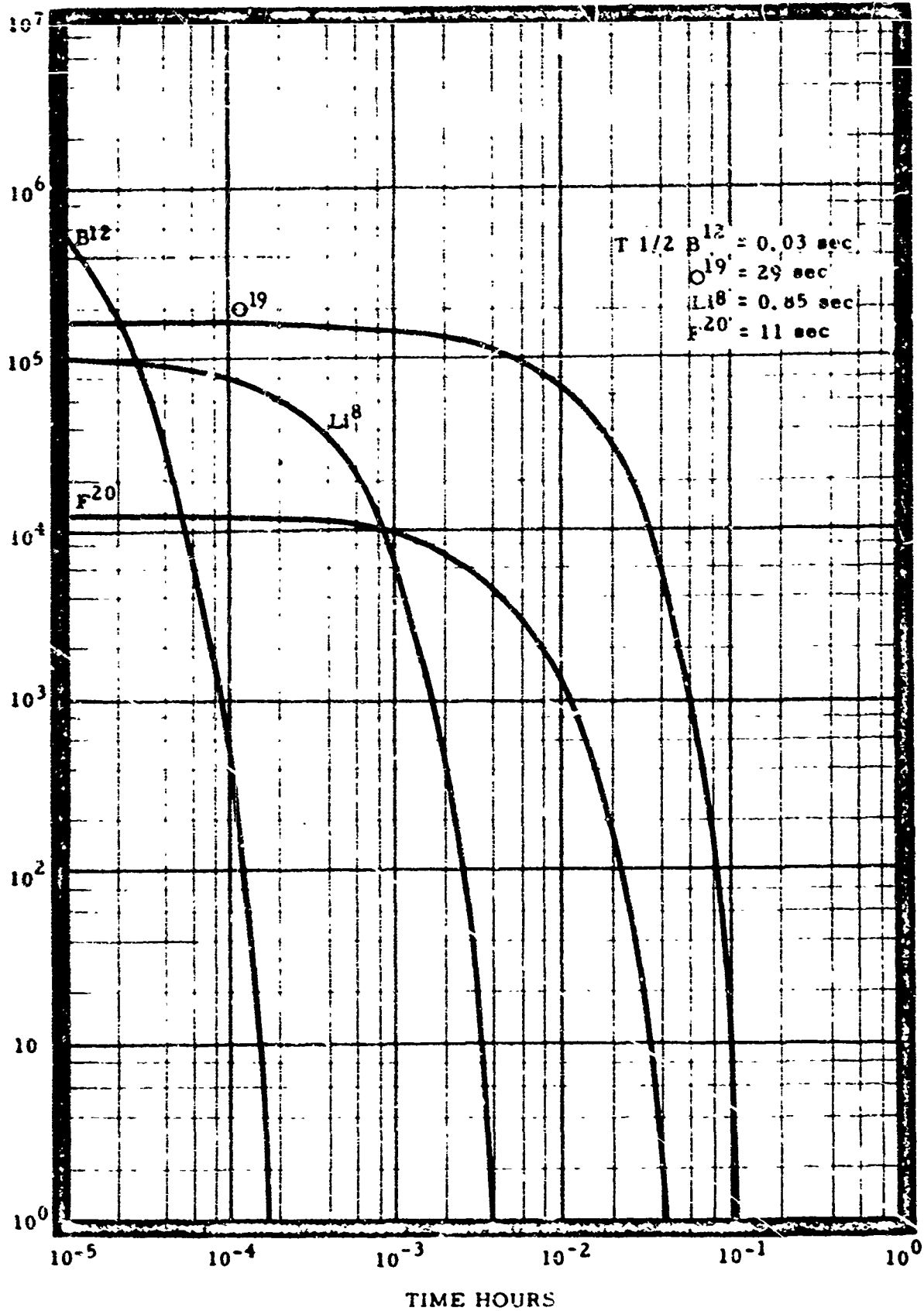


Figure 8 INDUCED ACTIVITY VS TIME (HALF LIVES OF LESS THAN 29 SECONDS)

Examination of Table 4 shows that effectively, two neutron absorbers, hydrogen and chlorine, account for most of the neutrons. Since hydrogen absorption is independent of the salt content, one can then write the approximation:

$$\sum_{i=1}^n C_i \cdot \sigma_{abs} \approx 35.856 + \frac{18.423 \cdot S}{3.45}$$

The total activity, A_{total} , then varies with the salt content according to:

$$A_{S \text{ total}} = A_{3.45 \text{ total}} \cdot \frac{S}{3.45} \left[\frac{54.986}{35.856 + \frac{18.423 \cdot S}{3.45}} \right]$$

We then have a convenient way of computing the total activity for any value of salinity once a given case has been computed carefully. The computed results are shown in Table 3 for 34.5 ‰ salinity.

The expression for the decay of the isotopes listed in Figure 7 is relatively straightforward with the exception of bromine-80. The activity as a function of time for the bromine-80 concentration is given by (Ref. 11):

$$A_2(T) = \frac{\lambda_1}{(\lambda_2 - \lambda_1)} \cdot A_1(0) \left[e^{-\lambda_1 T} - e^{-\lambda_2 T} \right] + A_2(0) e^{-\lambda_2 T}$$

where $A_1(0)$ is the activity of $\text{Br}^{80\text{m}}$ at $T=0$, expressed in units of curies.

$A_2(0)$ is the activity of Br^{80} at $T=0$, expressed in units of curies.

λ are the decay constant, expressed in units of seconds⁻¹

The pertinent values for $A_1(0)$, $A_2(0)$, λ_1 , and λ_2 are given in Table 4.

TABLE 6
SLOW NEUTRON INDUCED RADIOACTIVITIES IN SEA WATER

Active Isotope	Half Life	A (0) Curies	A(t) 1.2 hr	A(t) 1 hr	A(t) 15 mins	A(t) 10 min	A(t) 5 min	A(t) 1 min	A(t) 1 week	A(t) 2 weeks	A(t) 4 weeks	A(t) 12 weeks	A(t) 6 months	A(t) 1 year
^{60}Co	5.27 yr	1.37×10^{-9}												
^{36}Cl	3.08 yr	6.15×10^{-9}												
^{45}Ca	1.63 yr	5.92×10^{-8}												
^{14}C	5570 yr	9.25×10^{-3}												
^{90}Sr	12.46 yr	0.517												
^{60}Co	5.27 yr	2.1367	2.1365	2.1361	2.1322	2.1271	2.1209	2.1024	2.0724	2.032	1.9849	1.937	1.890	1.843
^{60}Co	5.27 yr	2.1357×10^3	2.1357×10^3	2.135×10^3	2.147×10^3	2.138×10^3	2.09×10^3	2.039×10^3	2.039×10^3	1.929×10^3	1.729×10^3	1.593×10^3	1.464×10^3	1.340×10^3
^{90}Sr	12.46 yr	3.078×10^{-2}												
^{90}Sr	12.46 yr	2.779×10^{-2}												
^{23}Na	25.1 day	0.2014												
^{32}P	14.3 day	3.432												
^{45}Ca	163 day	6.16×10^{-3}												
^{86}Br	16.3 hr	2.02×10^3	2.011×10^3	1.997×10^3	1.953×10^3	1.921×10^3	1.872×10^3	1.816×10^3	1.753×10^3	1.684×10^3	1.609×10^3	1.528×10^3	1.441×10^3	1.349×10^3
^{24}Na	15.03 hr	9.35×10^3	9.18×10^3	9.02×10^3	8.78×10^3	8.55×10^3	8.32×10^3	8.09×10^3	7.86×10^3	7.63×10^3	7.40×10^3	7.17×10^3	6.94×10^3	6.71×10^3
^{45}Ca	12.44 yr	2.38×10^3	2.338×10^3	2.294×10^3	2.250×10^3	2.206×10^3	2.162×10^3	2.118×10^3	2.074×10^3	2.030×10^3	1.986×10^3	1.942×10^3	1.898×10^3	1.854×10^3
^{90}Sr	12.44 yr	1.31×10^4	1.294×10^4	1.278×10^4	1.262×10^4	1.246×10^4	1.230×10^4	1.214×10^4	1.198×10^4	1.182×10^4	1.166×10^4	1.150×10^4	1.134×10^4	1.118×10^4
^{32}P	14.3 day	3.078×10^2	3.068×10^2	3.058×10^2	3.048×10^2	3.038×10^2	3.028×10^2	3.018×10^2	3.008×10^2	2.998×10^2	2.988×10^2	2.978×10^2	2.968×10^2	2.958×10^2
^{21}Na	2.02 hr	1.406	1.4118	1.418										
^{32}P	14.3 day	41.09												
^{35}Cl	37.29 min	6.94×10^5	3.97×10^5	2.28×10^5	10.87									
^{86}Br	16 min	6.79×10^5	2.23×10^5	7.96×10^4	2.64×10^4	8.32×10^3								
^{45}Ca	12.44 yr	1.397×10^3	1.4×10^3	1.43×10^3										
^{90}Sr	12.44 yr	8.39×10^3	7.29×10^3	6.31×10^3										
^{32}P	14.3 day	2.47×10^2	2.44×10^2	2.41×10^2										
^{45}Ca	12.44 yr	6.23×10^3	6.00×10^3											
^{90}Sr	12.44 yr	1.52×10^3												
^{32}P	14.3 day	1.237×10^4												
^{45}Ca	12.44 yr	7.91×10^2												
^{90}Sr	12.44 yr	1.03×10^5												
^{32}P	14.3 day	1.17×10^6												
TOTALS			5.129×10^5	3.39×10^6	5.46×10^6	2.619×10^5	7.43×10^3	2.147×10^3	1.93×10^3	2.73×10^3	2.10×10^3	1.39×10^3	8.3×10^2	1.39×10^3

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It will be noted that the first group of terms in the above equation shows the growth of the daughter (bromine-80) from the parent decay (bromine-80 metastable) and the decay of the daughter activity while the last term gives the contribution at any time from the initial daughter activity. (U)

4.2 THE NUCLEAR SUBMARINE AS A NEUTRON SOURCE

In this section we will consider the nuclear powered submarine as a source of neutrons and obtain an estimate of the induced activation in the surrounding sea water. Only the induced activities of sodium-24 and chlorine-38 are considered. (C)

The defining equation for activation analysis is given by (Ref. 12): (U)

$$A(t) = \sum_a \phi V(1 - e^{-\lambda T})_a - \lambda t \quad (\text{Eq. 1})$$

$$\text{where } \sum_a = \frac{2}{A} N_0 \sigma_a a_f \quad (\text{Eq. 2})$$

- $A(t)$ = disintegration rate for an arbitrary time, t .
- \sum_a = macroscopic activation cross section
- ρ = density
- A = mass number
- N_0 = Avogadro's number (6.025×10^{23})
- σ_a = microscopic activation cross section
- a_f = isotopic fractional abundance
- ϕ = neutron flux
- V = irradiated sample volume
- λ = decay constant
- T = irradiation time
- t = elapsed time after irradiation ceases

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In the following calculations the effective submarine hull area A from which neutrons escape is taken to be typically at 20 x 30 feet (the 20 foot dimension lies along the longitudinal axis of the submarine). The element of irradiated volume is taken as A times four diffusion lengths L , for thermal neutrons in water ($L = 2.85$ cm) (Ref. 13). The irradiation time T , is taken as the active length of the submarine hull (20 feet) divided by the submarine speed (20 knots). (C)

The analysis is certainly not for the general case since it has been tacitly assumed that the escaping neutrons are of thermal energy and those which are epithermal or fast will be moderated to thermal energy. This is not unrealistic since water is a relatively good moderator and the age-to-thermal for fission neutrons is known to be approximately 31 square centimeters (Ref. 14.) In addition a more general treatment must consider the neutron leakage as a function of the reactor power which in turn limits the submarine speed. (C)

The remaining terms of Equations 1 and 2 can be found in a number of good references on physical data. (U)

The results of the calculations are reported in Table 5. The effect of wake dilution has not been taken into consideration since no basis for an estimate could be obtained. However, if future wake dispersion studies indicate the amount of mixing in the wake so that a dilution factor X can be calculated, the concentrations would have to be decreased by this factor. The feasibility of detection can only be estimated without knowledge of this dilution factor. (C)

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TABLE 5. Neutron Flux and Induced Activity. (U)

Flux (φ) Neutrons /cm ² /sec	(N _T) Total (a) Neutron sec	PoBe (b) Equivalent (curies)	Total Initial Induced Activity Na ²⁴ + Cl ³⁸ (μc/ml)	Relative Concentration (c)
2 x 10 ³	1.1 x 10 ⁹	550	5.1 x 10 ⁻¹⁰	Below K ⁴⁰
10 ⁴	5.6 x 10 ⁹	2,800	2.55 x 10 ⁻⁹	Below K ⁴⁰
10 ⁵	5.6 x 10 ¹⁰	28,000	2.55 x 10 ⁻⁸	At K ⁴⁰
10 ⁷	5.6 x 10 ¹²	2,800,000	2.55 x 10 ⁻⁶	Above K ⁴⁰
10 ¹²	5.6 x 10 ¹⁷	2.8 x 10 ¹¹	0.255	Above K ⁴⁰

NOTE: (a) Effective hull area 600 square feet

(b) 2 x 10⁶ neutrons/sec/curie PoBe

(c) K⁴⁰ specific gamma activity 3 x 10⁻⁸ μc/ml (Ref. 16)

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Two limiting values of neutron fluxes, 2×10^3 and 10^{12} neutrons per square centimeter per second, were taken as lower and upper limits of expected possible values. The value of 2.5×10^4 neutrons per square centimeter per second has been experimentally determined (Ref. 15).

The polonium-beryllium equivalent is included in Table 5 in order to provide an indication as to the magnitude of the submarine as a neutron source. One curie values are not uncommon laboratory sources.

The feasibility column is a comparison between the natural potassium-40 concentration, a major background source, and the induced initial activity due to sodium-24 and chlorine-38.

Now it is interesting to know the activity of chlorine-38 relative to the activity of sodium-24 as a basis for possible tracking techniques. The defining equations can be seen to be independent of neutron flux, irradiated volume, or irradiation time.

$$A(t)_{Cl} = \sum_a^{Cl} V \phi (1 - e^{-\lambda_{Cl} t}) e^{-\lambda_{Cl} t} \quad (\text{Eq. 3})$$

$$A(t)_{Na} = \sum_a^{Na} V \phi (1 - e^{-\lambda_{Na} t}) e^{-\lambda_{Na} t} \quad (\text{Eq. 4})$$

It follows that

$$\frac{A(t)_{Cl}}{A(t)_{Na}} = \frac{(\sum_a \lambda)_{Cl}}{(\sum_a \lambda)_{Na}} e^{-(\lambda_{Cl} - \lambda_{Na}) t}$$

and with the proper values of

$$\sum_a^{Cl} = 4.54 \times 10^{-5} \text{ per cm} \quad \sum_a^{Na} = 1.482 \times 10^{-4} \text{ per cm}$$

$$\lambda_{Cl} = 3.1 \times 10^{-4} \text{ per sec} \quad \lambda_{Na} = 1.282 \times 10^{-5} \text{ per sec}$$

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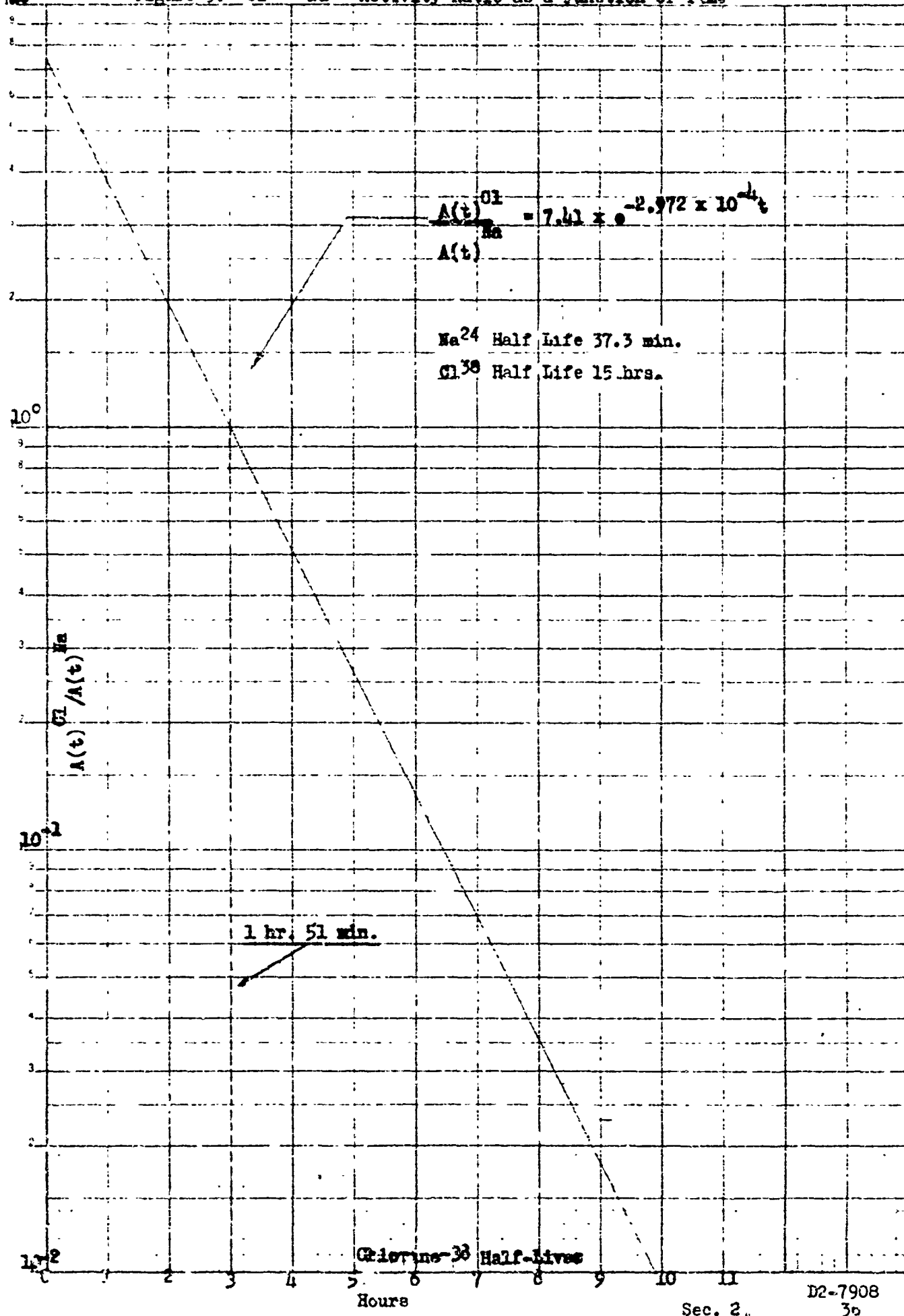
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$$\frac{A(t)_{Cl}}{A(t)_{Na}} = 7.41 e^{-29.72 \times 10^{-5} t} \quad (\text{Eq. 5})$$

Equation 5 has been plotted in Figure 9 as a function of chlorine-38 half-lives. It should be noted that after approximately 1 hour and 51 minutes, the chlorine-38 activity equals the sodium-24 activity. Figure 10 shows the decay schemes of these isotopes. (U)

Figure 9. Cl^{38} - Na^{24} Activity Ratio as a Function of Time



Cl^{38} Decay Scheme (Ref. 19)

$T_{1/2} = 37.29 \text{ Min}$

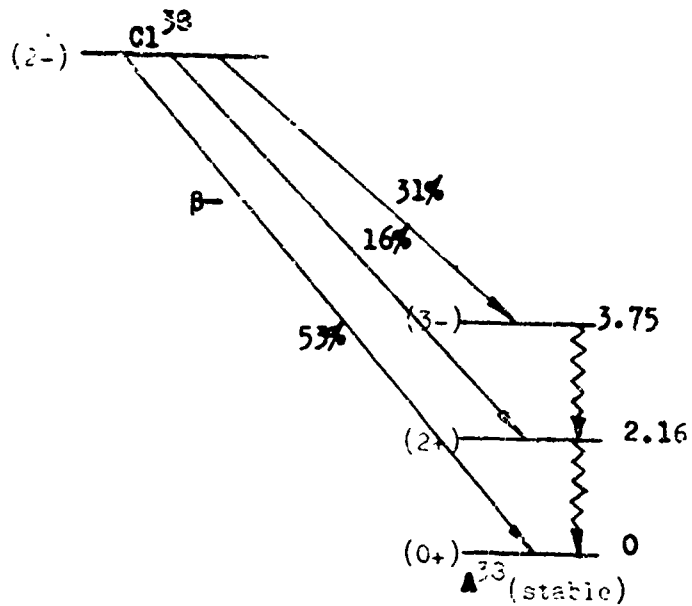
$\gamma_1 = 2.16 \text{ Mev}$

$\gamma_2 = 1.59 \text{ Mev}$

$\beta_{\text{max}}^- = 4.81 \text{ Mev (53\%)}$

$\beta_{\text{max}}^- = 2.77 \text{ Mev (16\%)}$

$\beta_{\text{max}}^- = 1.11 \text{ Mev (31\%)}$



Na^{24} Decay Scheme (Ref. 20)

$T_{1/2} = 14.97 \text{ hr}$

$\gamma_1 = 1.368 \text{ Mev}$

$\gamma_2 = 2.754 \text{ Mev}$

$\beta^- = 1.394 \text{ Mev}$

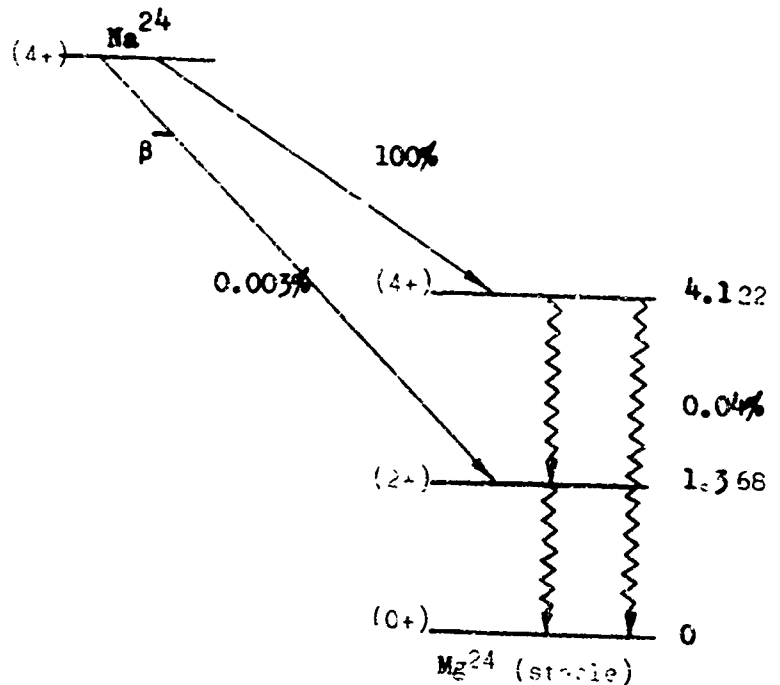


Figure 10. Relative Decay Schemes of Cl^{38} and Na^{24}

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5. COUNTING STATISTICS (Refs. 4, 17, 18)

In a practical counting experiment there are many factors which will contribute to the ultimate performance of a water monitor. The decay of a radioactive species is subject to statistical variation which is reasonably well understood. The problem is to detect weak concentrations of a given number of radioisotopes subject to statistical variations in the presence of a background which itself is subject to fluctuations.

A few relationships will be derived in this section which should be an aid to the reader in following some of the experimental results reported in the later sections.

5.1 MINIMUM DETECTABLE CONCENTRATION

The minimum detectable concentration, limited for the present only by statistical considerations, can be expressed as a function of several quantities.

Some principal quantities are:

1. Background counting rate.
2. Time duration of the observation period.
3. Counting efficiency of the instrument.
4. Confidence level.
5. Error one is willing to entertain.
6. Effective volume the detector "sees."

The necessary equations relating the above quantities have been largely worked out in a number of literature sources but are not

ideally suited, in the form presented, for ASW application (Refs. 4, 21). Therefore, the required expressions will be re-derived with some minor changes.

Consider the sea water to be contaminated with a concentration of radioactivity, C microcuries per milliliter ($\mu\text{C/ml}$), and let this activity result in a net sample counting rate of S counts per second (cps) being recorded by the instrument.

Let the average background contribute a counting rate of B cps.

The error, Y , of a counting-rate determination will be:

$$Y = K \sqrt{\frac{S+B}{T_s} + \frac{B}{T_b}}$$

where S is the net counting rate of the sample,

B is the counting rate of background alone,

T_s is the counting time of sample plus background,

T_b is the counting time of background alone,

K is the factor found in the third column of Table 6.

The minimum detectable signal is then:

$$S > K \sqrt{\frac{S+B}{T_s} + \frac{B}{T_b}}$$

where S is the counting rate due to the sample alone. This means, for example, that a counting rate of 4.9 ± 5.0 is not considered detectable, whereas 5.1 ± 5.0 is considered detectable.

TABLE 6. Summary of the Errors Most Used in Statistical Analysis of Radiological Counting Data

NOTE: n is the total number of observed counts, $n\bar{x}$ is the standard deviation excluding any contribution due to instrumental variability, and K is a factor relating standard deviation to probability.

Probability of Observing Error as Large as or Larger than Error Named in Column 1

Limits of Error in Radiochemical Counting

Name of Error	in Column 1	$K \bar{x}$ std. dev.	Remarks
Probable Error	0.500	$0.6745(n)^{1/2}$	Quite commonly used in scientific work of all kinds. Becoming obsolete from a statistical standpoint. One half of the values in a series of similar counting determinations will be in error by less than the probable error.
Standard Deviation	0.3173	$1.000(n)^{1/2}$	Most used statistically since most statistical tables are computed as functions of the standard deviation. Usually symbolized as the Greek letter sigma.
Nine-tenths Error	0.1000	$1.645(n)^{1/2}$	Commonly used in radiocounting. Named because there are nine chances out of ten that the error will be smaller. Sometimes abbreviated N.T.E. Statisticians would call this the 0.10 level of significance but would not consider deviations with a probability as high as this very significant.
Ninety-five Hundredths Error	0.0500	$1.96(n)^{1/2}$	Most commonly used by statisticians as the level at which deviations exceed chance variation and thus are considered significant. Called the 0.05 level of significance.
Ninety-nine Hundredths Error	0.0100	$2.576(n)^{1/2}$	Deviations of this magnitude may be considered highly significant. Often used by statisticians and called the 0.01 level of significance.

The fractional error, F , can be written as:

$$F = \frac{Y}{S} = \frac{K \sqrt{\frac{S+B}{T_s}} + \frac{B}{T_b}}{S}$$

Now to minimize the error by efficient distribution of counting times:

$$\frac{S+B}{B} = \left(\frac{T_s}{T_b} \right)^2$$

Suppose for a low-level counting experiment that $S+B \approx B$, then it follows that $T_s = T_b = T$

The expression for the fractional error can then be replaced by

$$F = \frac{K \sqrt{\frac{S+2B}{T}}}{S}$$

or

$$S = \frac{K^2 + K \sqrt{K^2 + 8F^2 \cdot T \cdot B}}{2F^2 \cdot T} \text{ counts per second}$$

The efficiency of the instrument, E , is defined as the ratio of the recorded counting rate to the disintegration rate that occurred in the sample during one counting interval.

Symbolically then

$$E = \frac{S \text{ counts per sec}}{3.7 \times 10^4 \frac{\text{disT}}{\text{sec}} \times \frac{C \mu\text{C}}{\text{ml}} \times V \text{ ml}}$$

microcuries

where V is the volume expressed in units of milliliters.

C is the concentration expressed in units of microcuries per milliliter.

Substituting for S in the last equation and solving for C yields:

$$C = \frac{K^2 + K \sqrt{K^2 + 8F^2 \cdot T \cdot B}}{7.4 \times 10^4 F^2 \cdot E_s \cdot V_s \cdot T} \mu\text{C/ml}$$

Now if the background is predominantly due to some radioisotope in the sample (or the volume of sea water seen by the detector) then:

$$B = \frac{\text{dist/sec}}{\text{ml}} \times V_s \text{ ml} \times E_s \frac{\text{counts/sec}}{\text{dist/sec}}$$

If there are a number of volume distributed background sources, then:

$$B = \sum_{i=1}^n B_i V_{i6} E_i$$

In addition there may be contributions to the background which are known only in terms of B_j counts per second. The total background contribution is given:

$$B = \sum_{i=1}^n B_i V_{i6} E_i + \sum_{j=1}^m B_j$$

Then:

$$C = \frac{K^2 + K \sqrt{K^2 + 8 F^2 T \left(\sum_{i=1}^n B_i V_{i6} E_i + \sum_{j=1}^m B_j \right)}}{7.4 \times 10^4 F^2 E_s V_s T}$$

5.2 COMPUTER STUDY

A study is underway in which the pertinent parameters are permuted in meaningful combinations which together with the experimental data should provide a sound basis for detector design. The study is being performed with the aid of an IBM 704 electronic computer.

One of the objectives of this study is to accumulate data which will facilitate a choice of detectors. In Section 7.2 of this report data is presented for different phosphors of comparable dimensions. Each detector will have a net sample counting rate and a background rate. The following paragraphs develop the analytical expressions involved in the comparison of detection systems.

5.3 THRESHOLD BACKGROUND

An improvement in counting statistics implies that a counting rate of 5.0 ± 2.0 cpm is poorer than 5.0 ± 1.0 cpm or 50 ± 12 cpm.

Symbolically then:

$$\left(\frac{K\sigma_1}{S_1}\right) > \left(\frac{K\sigma_2}{S_2}\right)$$

It follows that:

$$\frac{B_2}{B_1} < \frac{\left(\frac{S_2}{S_1}\right)^2 \times \left(\frac{2B_1}{S_1} + 1\right) - \frac{S_2}{S_1}}{2 B_1/S_1}$$

The proportionate change in background counts, B_2/B_1 , can be regarded as the dependent variable and the proportionate change in sample counts S_2/S_1 , as the independent variable with the original background to sample counting ratio, B_1/S_1 , as the parameter in the above inequality. Figures 11 and 12 are plots of this inequality.

Two examples are offered as an aid in using these graphs:

1. For a decrease in the net sample counting rate from S_1 to S_2 , what is the minimum change in background counting rate, B_1 decreases to B_2 , so that the statistics will be conserved? Let the ratio S_1/B_1 , be 0.5 and assume a change in sample count rate S_2/S_1 of 0.4. Taking these values to Figure 11, B_2/B_1 is 0.1 which means that the background must be reduced at least to one tenth of its original value if statistical reliability is to be conserved.
2. For an increase in net sample counting rate, let S_2/S_1 be 2 and S_1/B_1 be 5. From Figure 12 it is seen that the increase in background, B_2/B_1 , may be as much as 9 with no loss in statistical reliability.

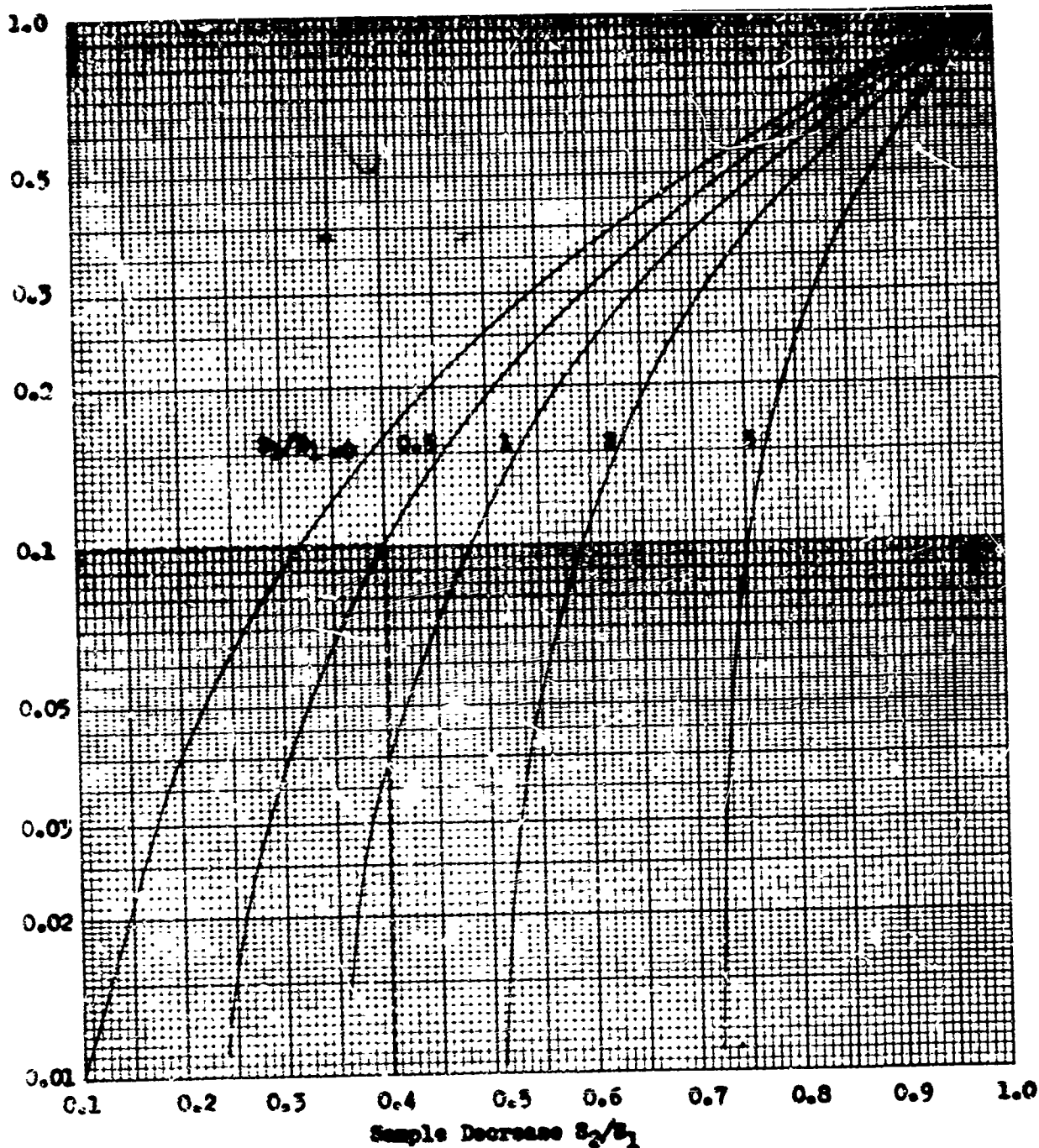
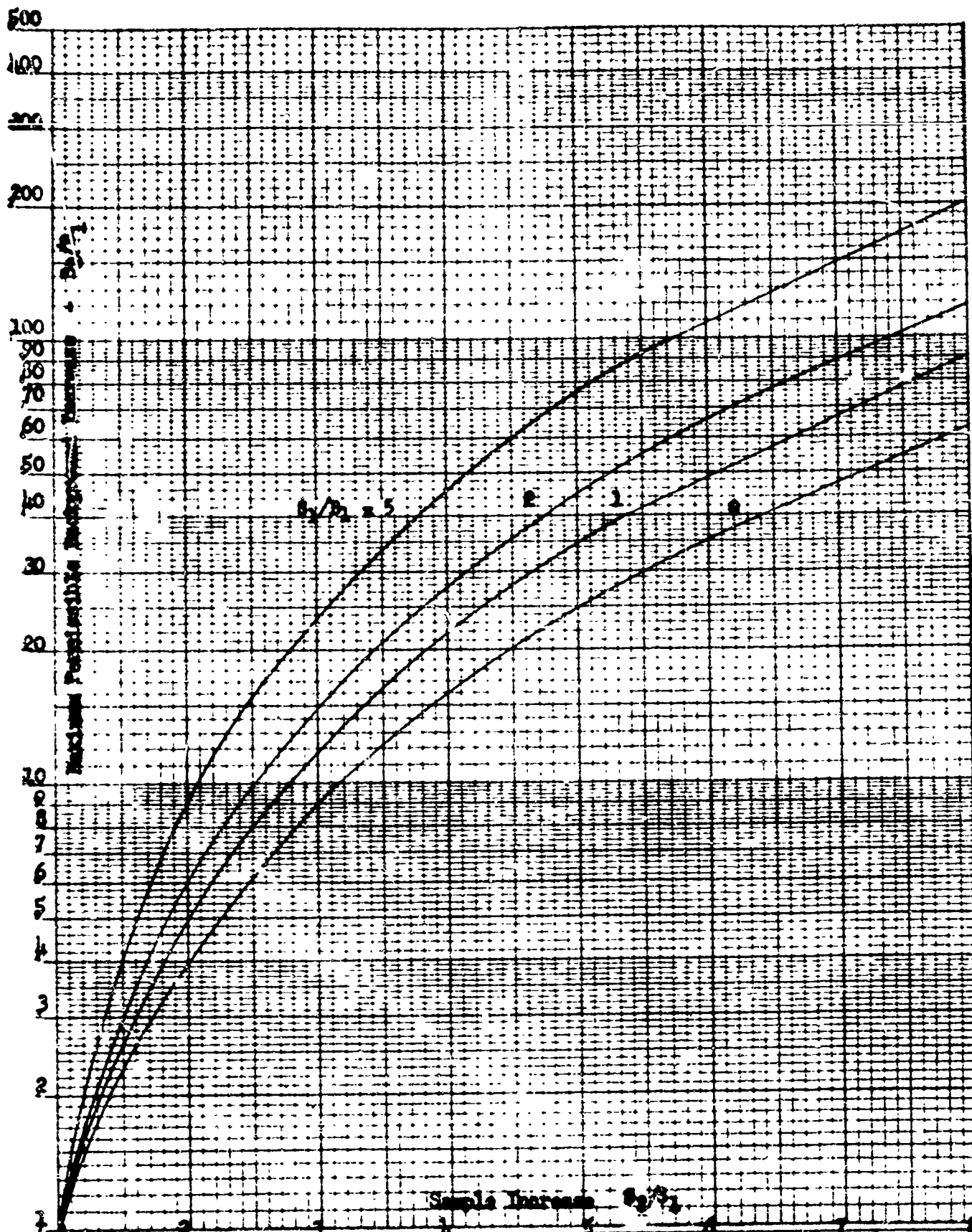


Figure 11. Required Decrease in Background Counting Rate in order to Retain Statistical Reliability when there is a Decrease in the Sample Counting Rate.



CALC CHECK APR APR		REVISED	DATE	Figure 12. Allowable Increase in Back-ground for a specified Increase in Sample Counts. BOEING AIRPLANE COMPANY SEATTLE 24, WASHINGTON	D2-7908
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Consider now the following situation. Suppose we have an initial net signal to background ratio of S_1/B_1 and that it is possible to reduce the background to essentially zero but in doing so the net signal is also reduced. This problem comes up in background depression. The question is what level S_2 is required for $B_2 = 0$ so that the statistics are preserved for a given counting time? The necessary expression is seen to be:

$$\text{or: } \frac{\left(\frac{S_2}{S_1}\right)^2 \left(\frac{2B_1}{S_1} + 1\right) - \frac{S_2}{S_1}}{2B_1/S_1} > 0 \quad B_2 \approx 0$$

$$\frac{S_2}{S_1} > \frac{1}{2\left(\frac{B_1}{S_1}\right) + 1}$$

This inequality is graphically portrayed in Figure 13. The physical significance of this last inequality is that reducing the net sample counting rate S_2 below the value required by the inequality will result in poorer counting statistics or a longer counting time to preserve the statistic, even though the background has been eliminated.

Example:

Let the count rate from sodium-24 in a raw sample of sea water be $S_1 = 200$ cpm.

Let the potassium-40 background be $B_1 = 200$ cpm so that

$$\frac{S_1}{B_1} = 1.0$$

Now say that all the potassium-40 is removed so that $B_2 = 0$ what must the sodium-24 counting rate be so that the statistics remain invariant for a given counting time?

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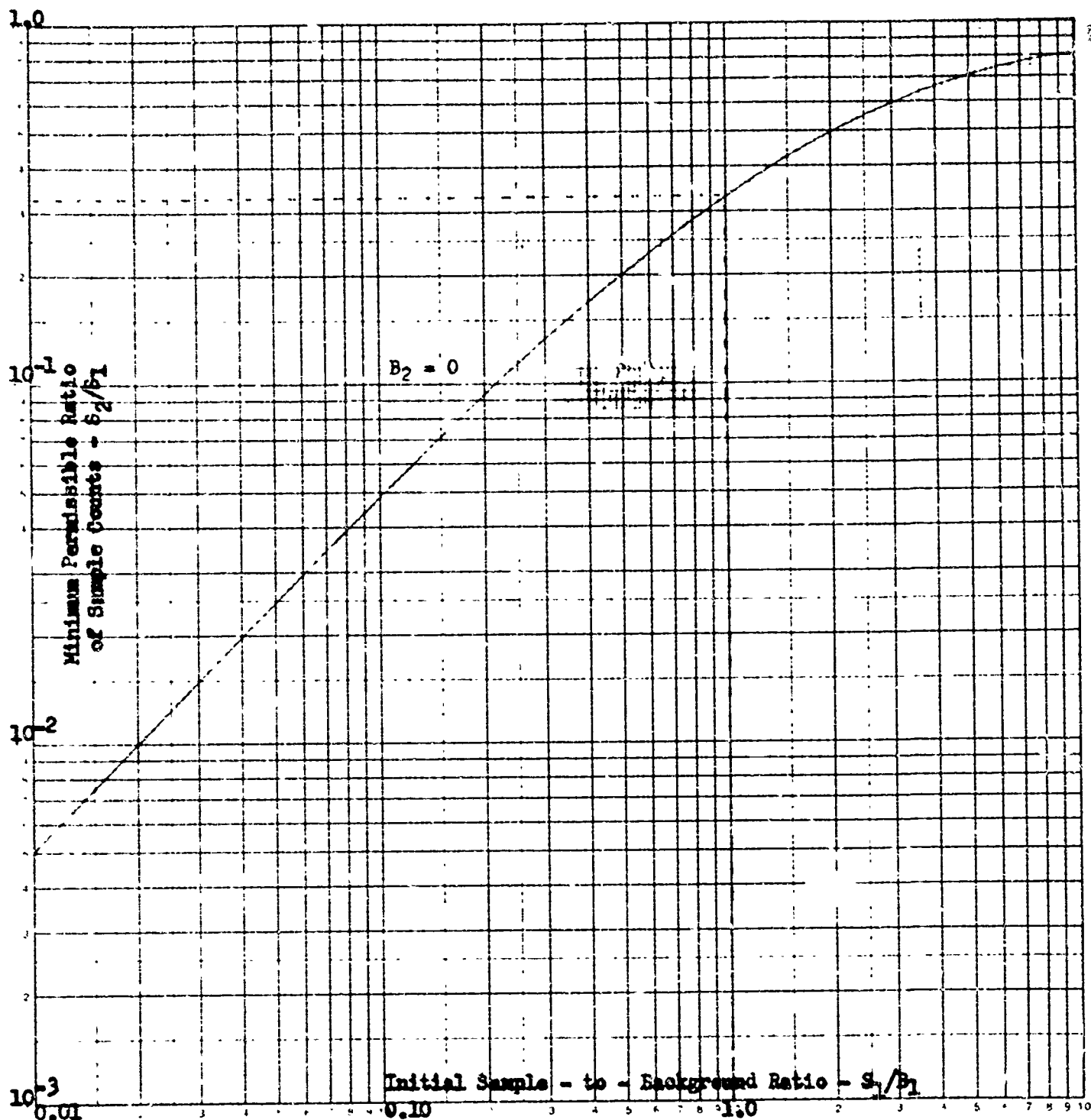


Figure 13. Allowable Reduction in Sample counts when Background is Eliminated. Based on initial Sample to Background ratio.

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From Figure 1:

$$S_2 = 0.33 \times S_1 = 0.33 \times 200$$

$$S_2 = 66 \text{ cpm}$$

That is, with all the potassium-40 removed the sodium-24 must yield a counting rate of 66 cpm in order to preserve the statistics when compared to an initial sodium-24 count rate of 200 cpm and a potassium-40 count rate of 200 cpm.

This example assumes that the sole contributor to the background is potassium-40 content in the sea water. This is not so but serves to illustrate the idea. This fact should be kept in mind considering the potassium-40 background suppression discussion under the chemical concentration section.

5.4 COUNTING TIME

In an effort to establish an estimate of the counting times required to satisfy a specified confidence level and fractional error, data collected in the laboratory were used in the following equation:

$$t = \frac{K^2}{F^2} \frac{1}{S^2} (S + B)$$

where t = counting time

K = factor from column 3, Table 6

F = fractional error

S = net sample counting rate

B = background counting rate

The above equation assumes that the background counting rate will have been well established prior to the experiment and its error small compared to the magnitude of the signal.

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The counting times which would be required to detect the induced activity in the wake of a nuclear-powered submarine are presented in Figures 14 to 17 as functions of the total induced activity in the wake. These curves were obtained from the predicted spectral shape in a 5 x 4 inch NaI(Tl) crystal for the induced activity produced by the flash activation of the sodium and chlorine in sea water immediately after the passage of the submarine. Integral counting techniques were assumed in that all counts above a given energy were counted. Two values were taken for the lower energy level (100 kev and 1.6 Mev). The counting times which are needed depend very sensitively on the statistical certainty of detection which is desired. Curves are given for a confidence level of 95% with a fractional error of 5 percent and for a confidence level of 50 percent with a fractional error of 50 percent. (C)

Two choices for the background were made. In the first case the background was assumed to be due solely to the sea water concentration of potassium-40; and the second case, for a background which included, in addition to the potassium-40, the background which was measured in our tank filled with tap water. The background spectra which were used are shown in Figures 5 and 6. The actual background, hence the counting times, will be intermediate between the two sets of curves. (U)

In Table 8, page 10⁶, will be found the approximate neutron leakage fluxes which would produce the corresponding specific activities for a submarine speed of 20 knots and assuming no dilution of the wake. (C)

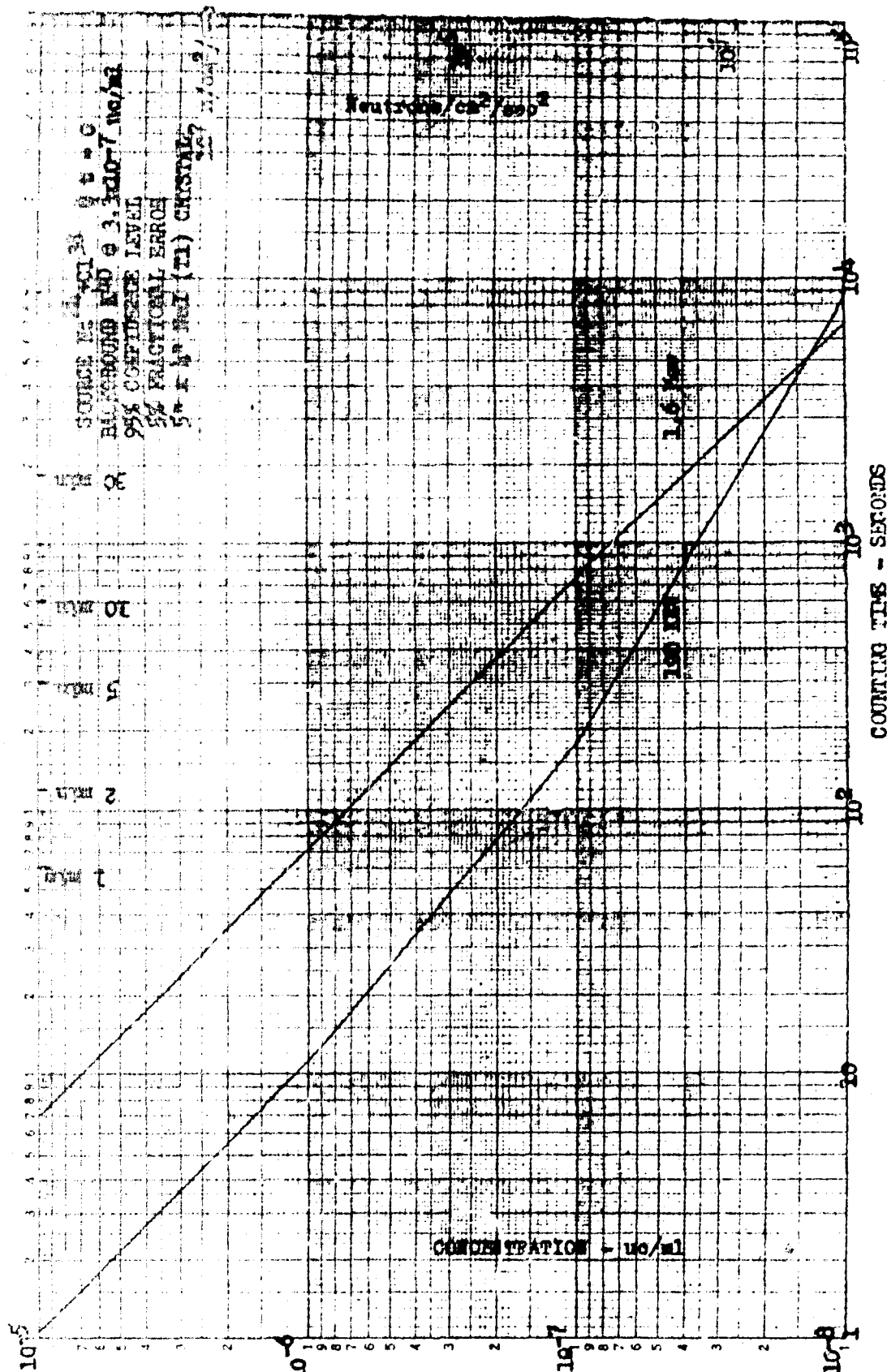
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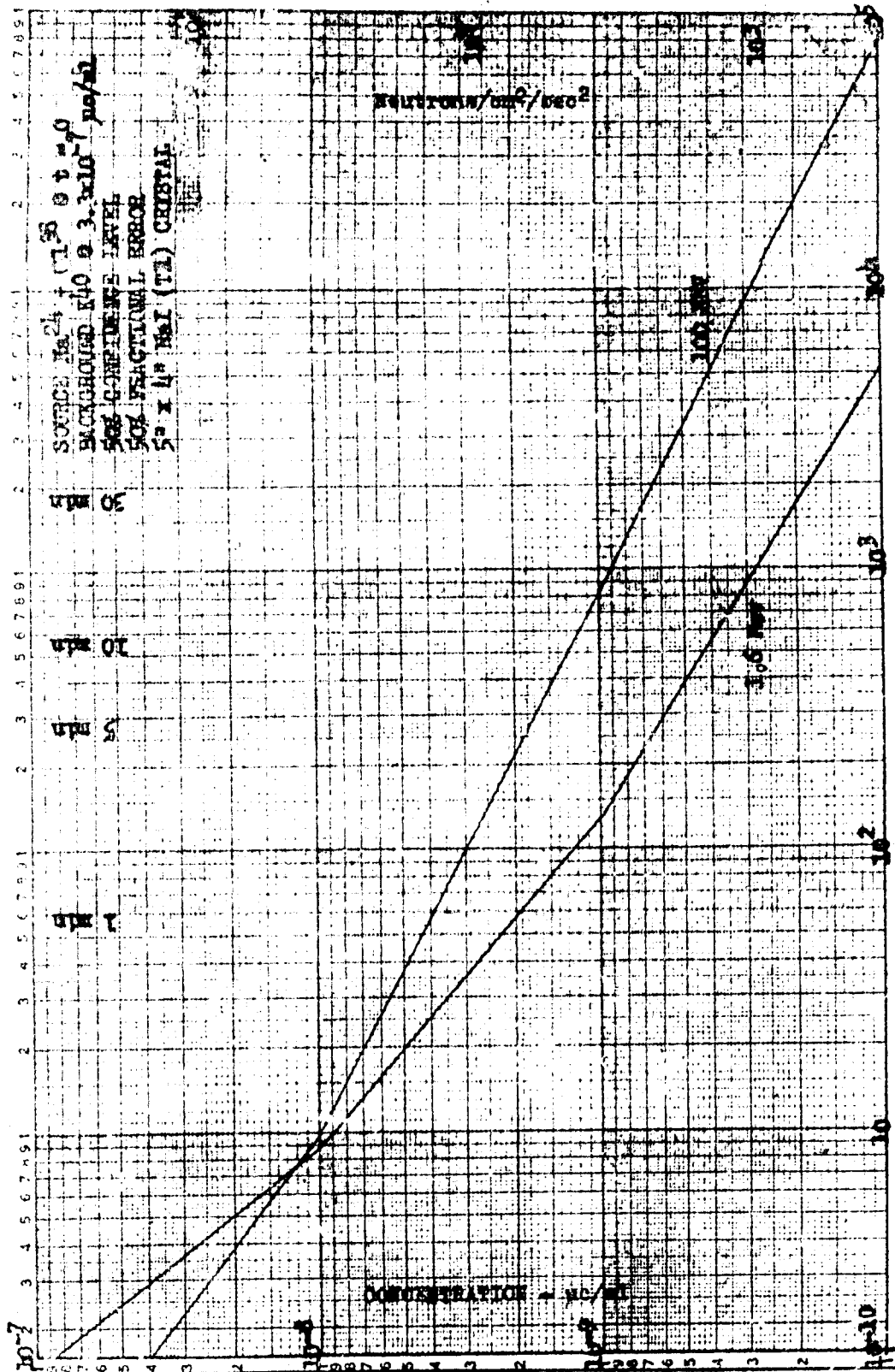
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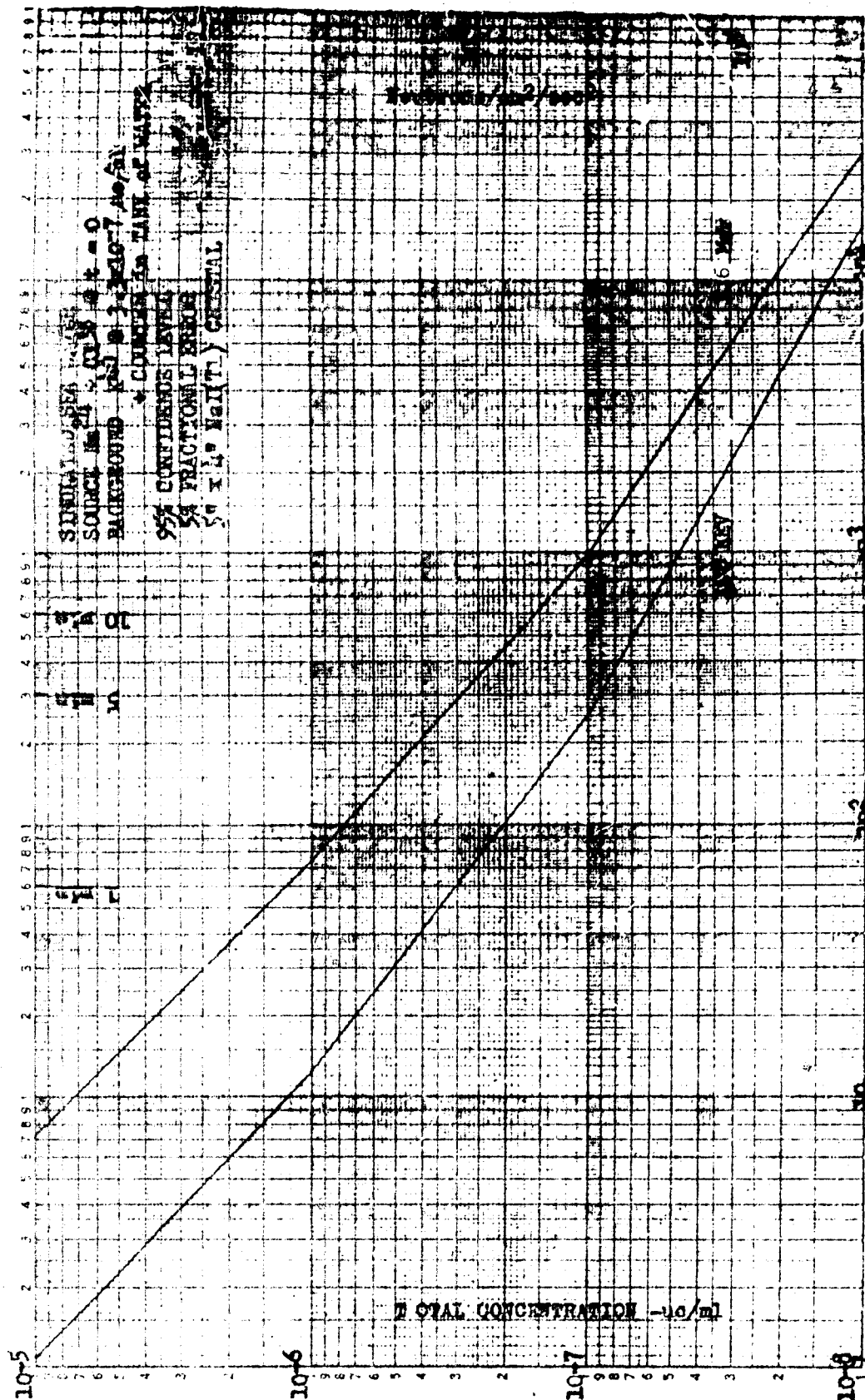


REVISED	DATE	Figure 14. Sample Concentration vs. Counting Time with ^{240}Pu Background, 95% Confidence Level.	D2-7908
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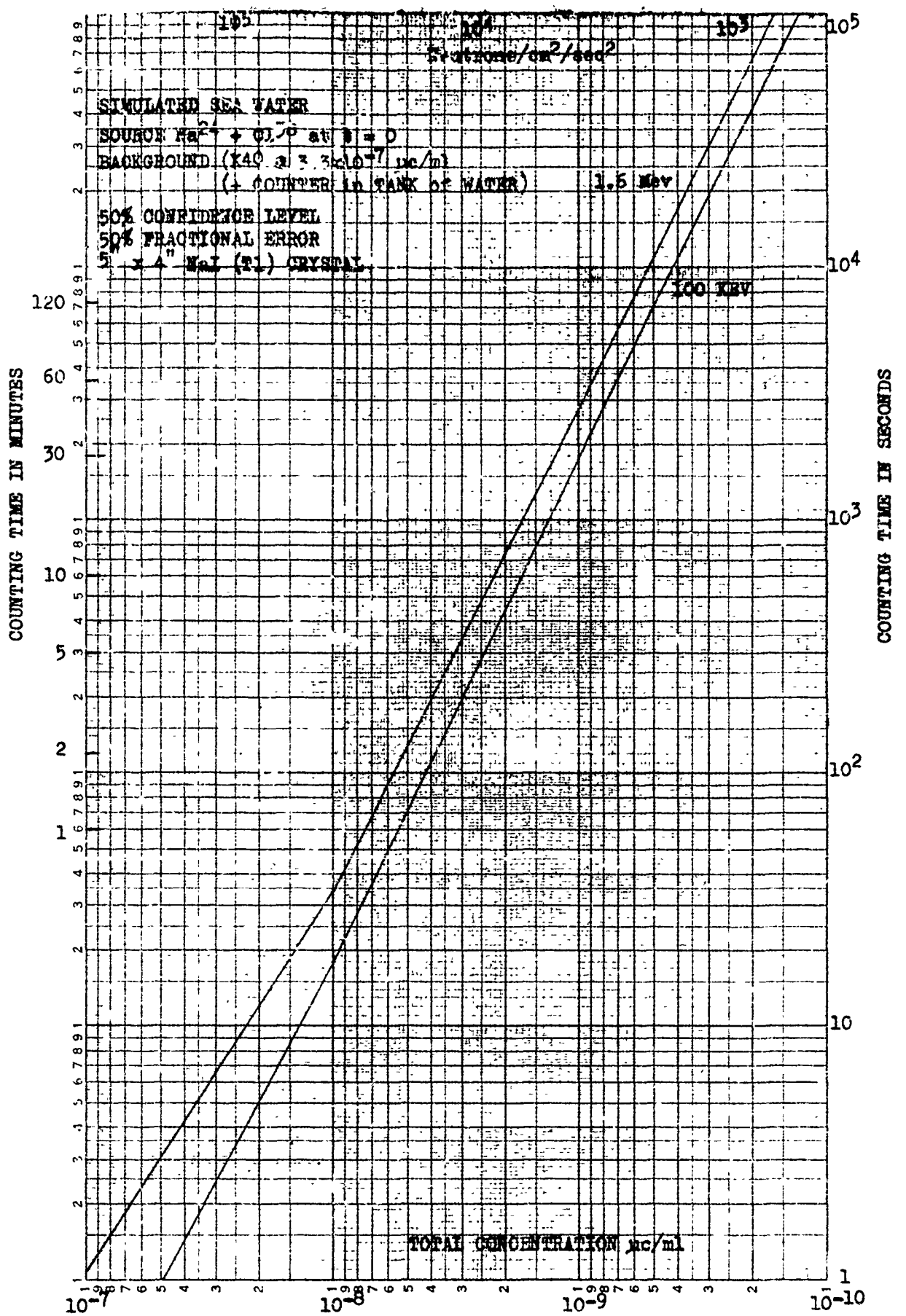


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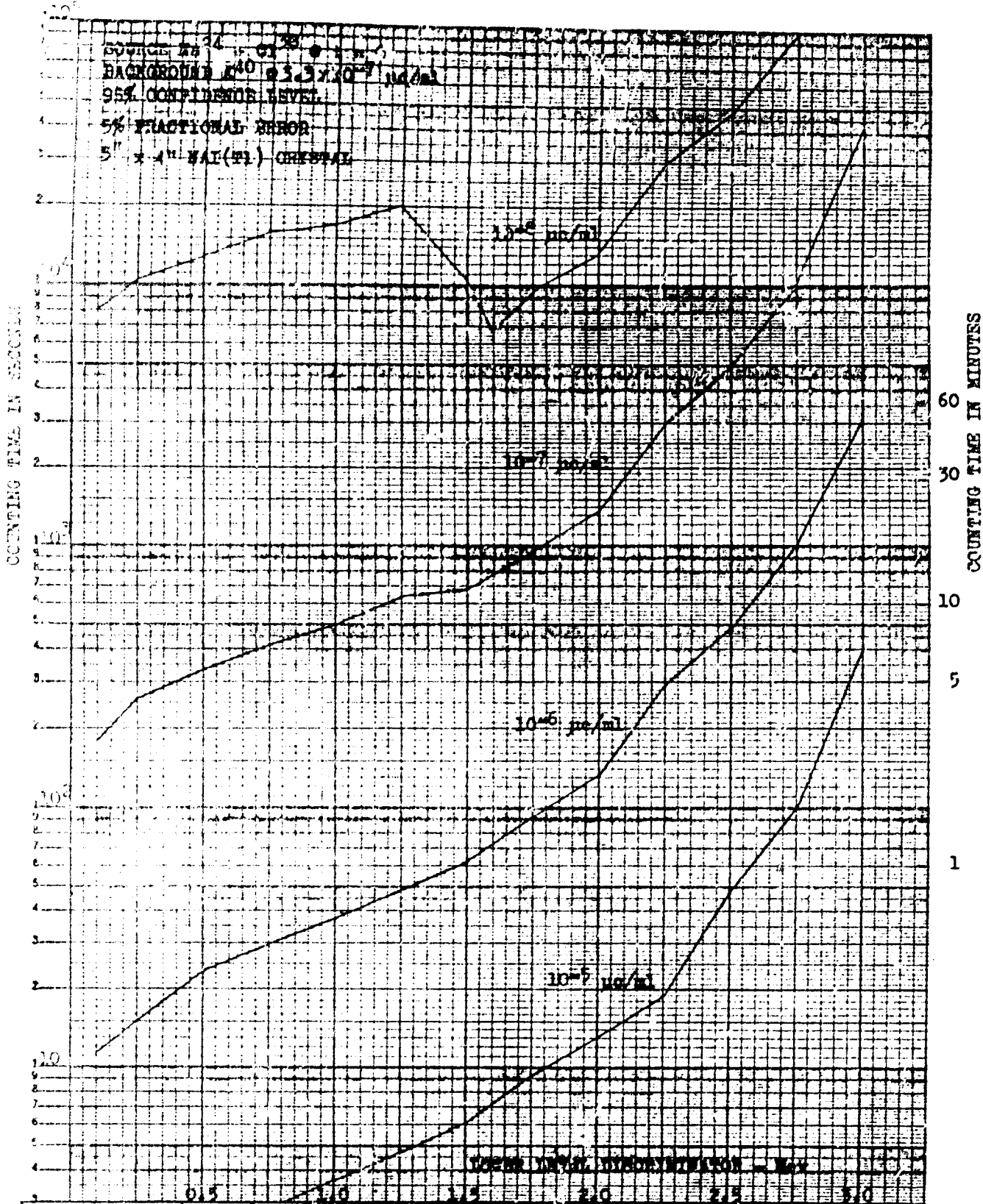
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	REVISED DATE	Figure 16. Sample Concentration vs. Counting Time With K^{40} and Tank Background, 95% Confidence Level BOEING AIRPLANE COMPANY	D2-7900 Sec. 2. PAGE 52
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CALC		REVISED	DATE	Figure 17. Sample Concentration vs. Counting Time With K^{40} and Tank Back- ground, 50% Confidence Level BOEING AIRPLANE COMPANY SEATTLE 24, WASHINGTON	D2-7908
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The effect of energy discrimination is more clearly seen in Figures 18 to 21 where the required counting times are plotted as functions of the lower level setting for various values of the specific activity of the induced activity.



REVISED	DATE	Figure 18. Counting Time vs. Lower Level Discriminator Setting With K^{40} Background, 95% Confidence Level BOEING AIRPLANE COMPANY SEATTLE 24, WASHINGTON	D2-7908 Sec. 2. PAGE 55 OF
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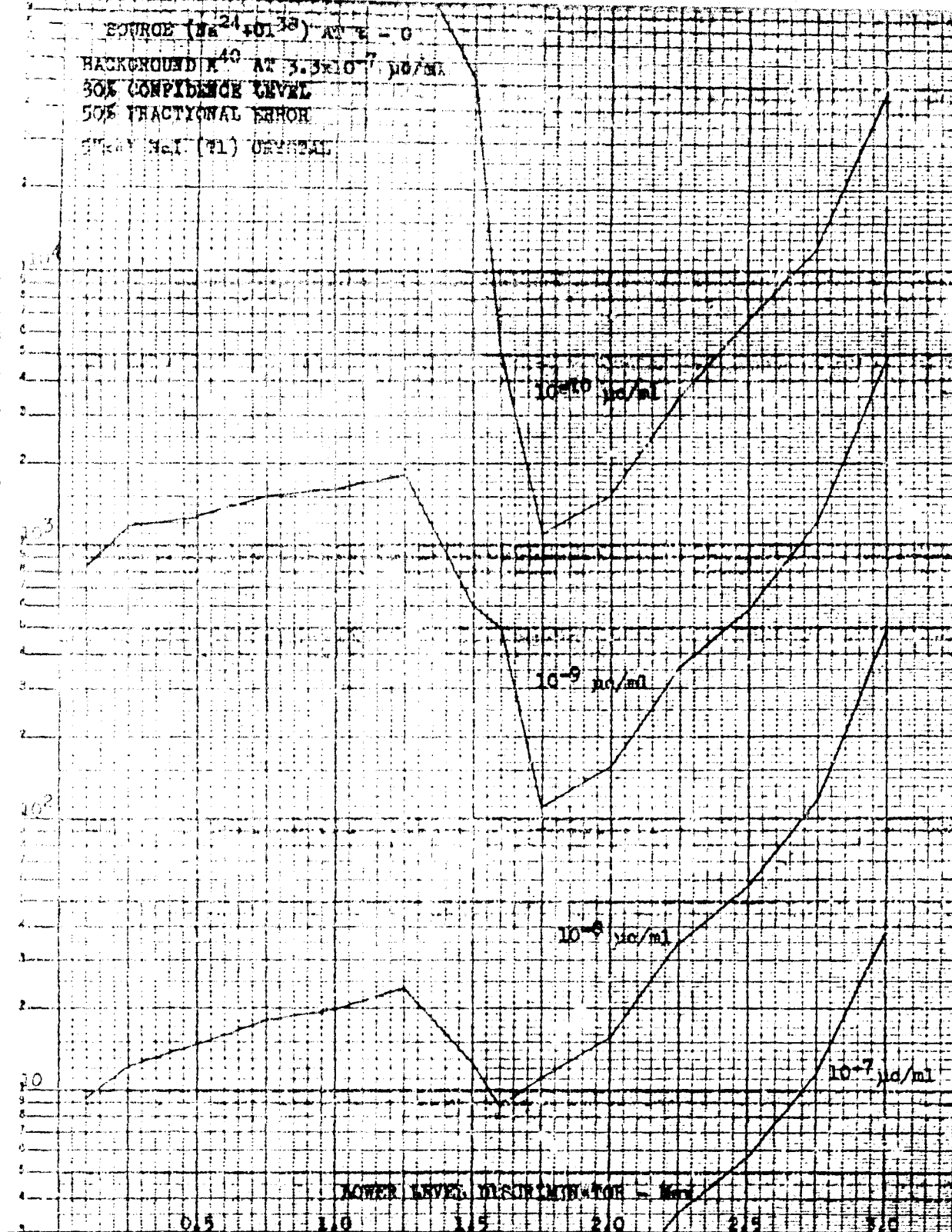
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COUNTING TIME IN SECONDS

COUNTING TIME IN MINUTES



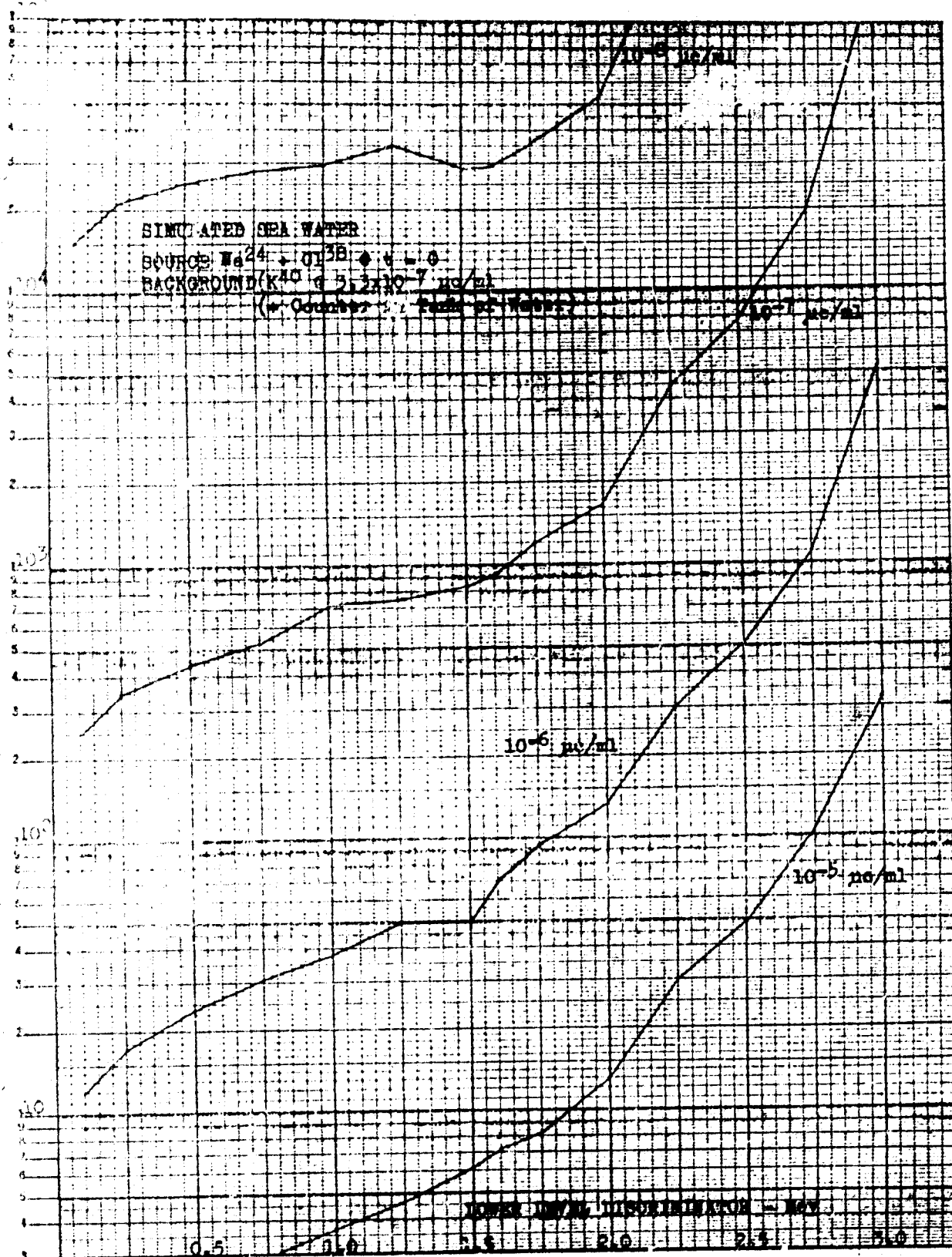
REVISED	DATE	Figure 19. Counting Time vs. Lower Level Discriminator Setting With K^{40} Background; 50% Confidence Level	D2-7903
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COUNTING TIME IN SECONDS

COUNTING TIME IN MINUTES



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Figure 20. Counting Time vs. Lower Level Discriminator Setting With K40 and Tank D2-7908 Background, 95% Confidence Level

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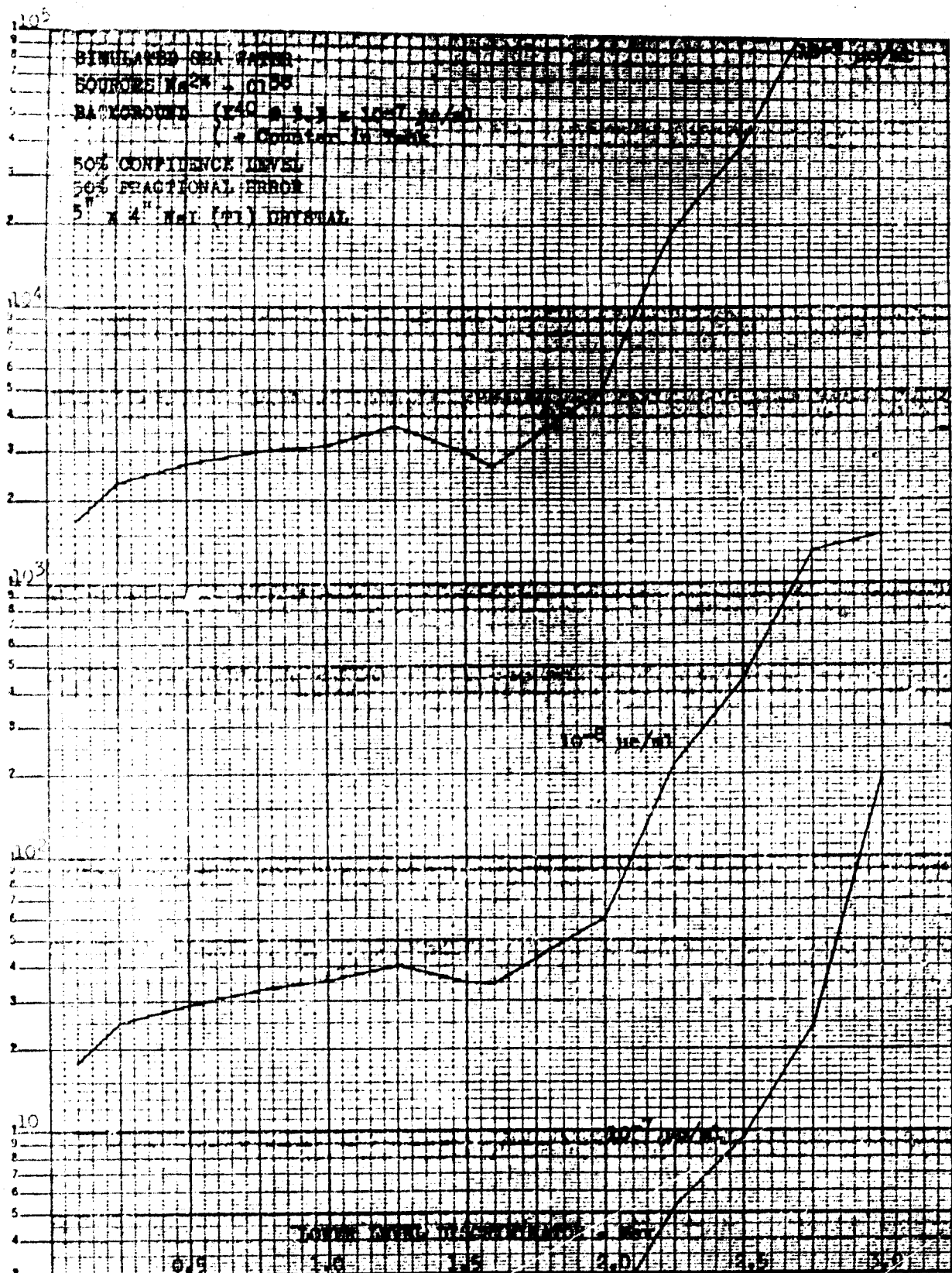
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COUNTING TIME IN SECONDS

COUNTING TIME IN MINUTES



DATE	REVISED	DATE	Figure 21. Counting Time vs. Lower Level Discriminator Setting With ^{40}K and Tank Background, 50% Confidence Level BOEING AIRPLANE COMPANY SEATTLE 24, WASHINGTON	D2-7908 Sec. 2. PAGE 53 OF
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6. EXPERIMENTAL PROCEDURE

6.1 INTRODUCTION

Volume distributed spectra were obtained for solutions of potassium-40, sodium-24^{and} chlorine-38. These isotopes correspond to the chief contributors to the radioactive background in sea water (potassium-40) and the principal radioisotopes produced by the submarine (sodium-24 and chlorine-38). The experimental data was obtained as follows: a waterproof detector was suspended at the center of a large cylindrical tank containing a mixture of one of the isotopes and water. Pulses from the detector were fed to a 256-channel analyzer, and the spectrum was recorded. The tank will be shown in Figure 29, page 72.

6.2 SOURCES

The potassium-40 was obtained from commercial grade (muriate of potash) potassium chloride. A chemical assay of the material showed it to contain 95.63 percent potassium chloride. Since 0.012 percent of natural potassium is potassium-40, a specific activity of 2.26×10^{-6} $\mu\text{C}/\text{ml}$ was obtained in the tank for each 100 pounds of the potassium chloride. Approximately 200 pounds were used for each series of measurements.

The sodium-24 and chlorine-38 were produced by neutron bombardment in the University of Washington 60-inch cyclotron. The neutrons were obtained by the bombardment of a beryllium plate by 20 Mev deuterons in the target chamber of the cyclotron. Previous measurements by cyclotron personnel have shown that fast neutron fluxes of about 10^{10} neutrons per square centimeter per second were obtained with deuteron beams of about 100 microamperes. Most of the cyclotron runs were with

beams of approximately 120 microamperes for a period of about one hour.

The sodium-24 was made by placing approximately 225 grams of sodium chloride in small bottles directly in the fast neutron flux (Figure 22). The measurements were made in the tank on the day following the bombardment so that the induced chlorine-38 activity had decayed to an insignificant level.

The fast neutron flux could not be used to produce the chlorine-38 because of the simultaneous production of chlorine-34 by the $(n, 2n)$ reaction with chlorine-35. Chlorine-34 has a half-life of about 32 minutes very close to that of chlorine-38 (~ 37 minutes), and a variety of gamma rays with energies which would interfere with the determination of the chlorine-38 spectrum. The fast neutron flux was therefore moderated by placing the sample to be bombarded in a 2-inch diameter hole in a cylindrical container (approximately 22 inches in diameter) of paraffin. The container and sample were then placed as close as possible to the cyclotron target chamber (Figure 23). This arrangement reduced the amount of chlorine-38 activation by about an order of magnitude, but with a negligible amount of chlorine-34 contamination. About 225 grams of lithium chloride was bombarded at a time to obtain chlorine-38 samples free of sodium-24 activity. The induced activities in the lithium were of very short half-life and had decayed before the counting was performed. The specific activities of the sodium-24 and chlorine-38 solutions were in the range from one to ten microcuries per milliliter.

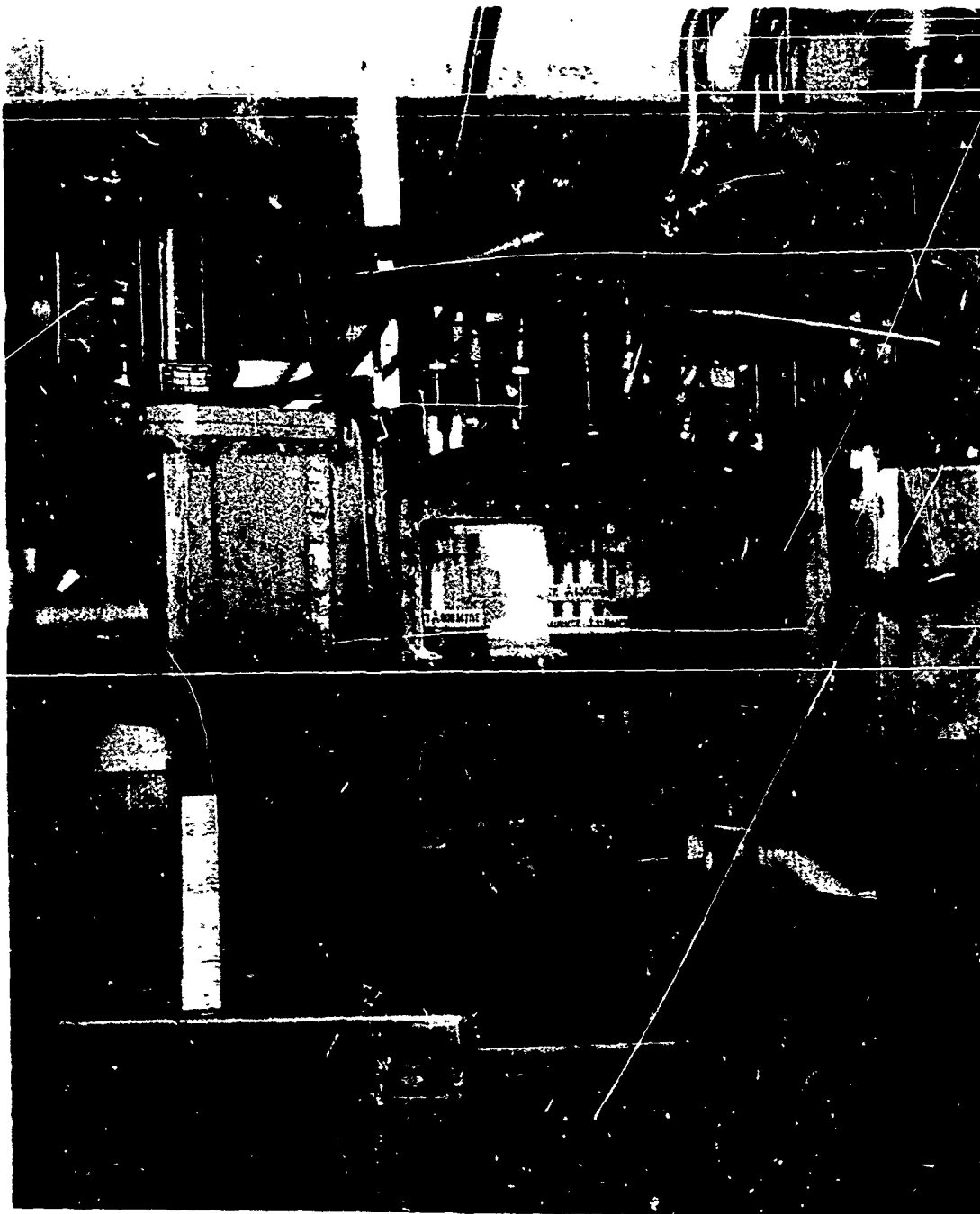


Figure 22. Cyclotron Target Box with Racks Containing Sample Vials. A beam of 20 Mev deuterons enters from the right and strikes a beryllium plate inside the target box. High energy neutrons emitted by the beryllium, irradiate the NaCl samples (in racks, center of photo). A few hours are allowed for the decay of Cl^{34} and Cl^{38} (both about 1/2 hours half life) after which Na^{24} is the only detectable gamma emitter.

The paraffin block, shown between the sample racks, was ineffective as a moderator (see Figure 23).

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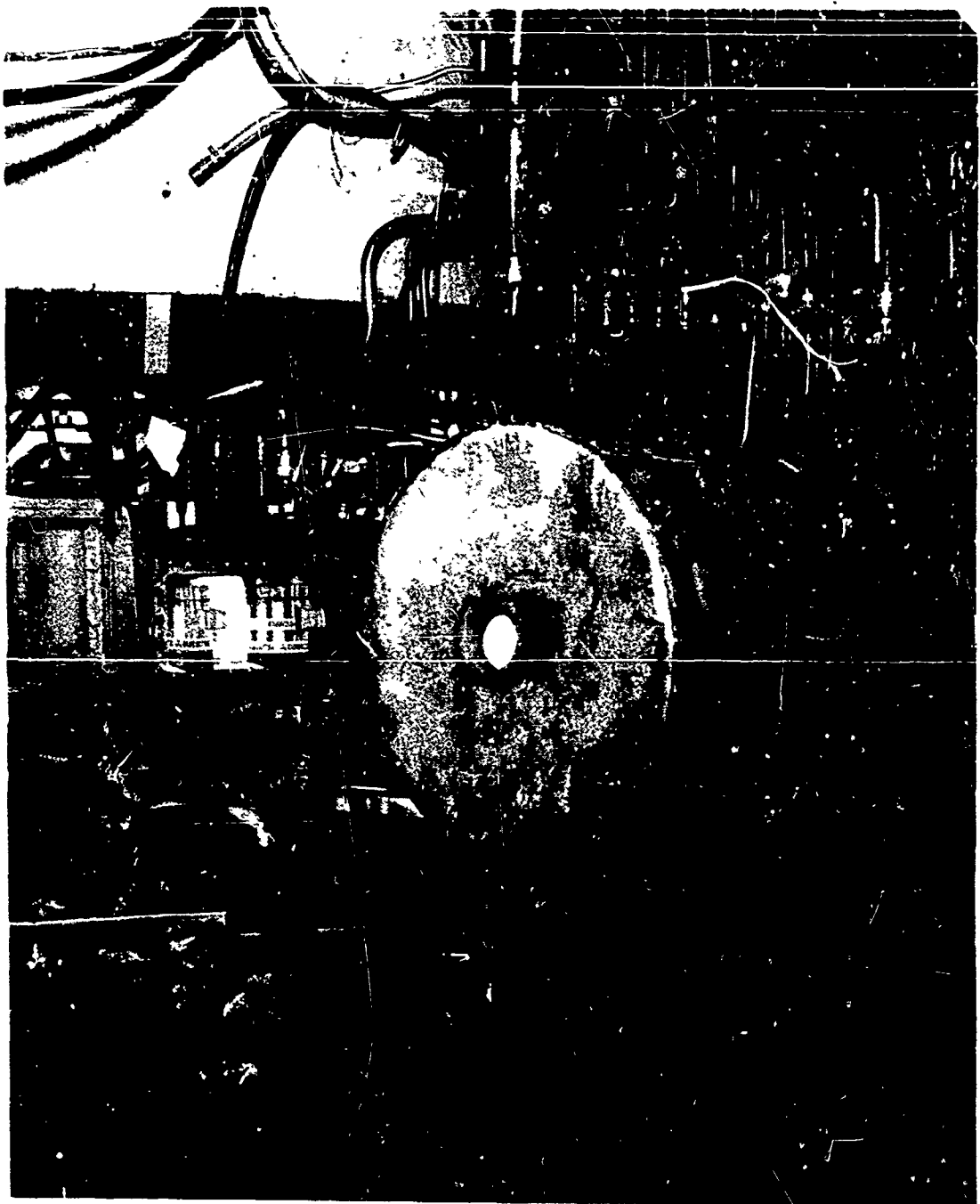


Figure 23. Paraffin Moderator for Preparation of C^{138} . Lithium chloride samples are placed in the center of the large, cylindrical container where they are surrounded by about 10 inches of paraffin. Scattered, high energy neutrons from the beryllium target are moderated by the paraffin to below the $(n, 2n)$ energy threshold for production of C^{134} so that only C^{138} is produced in the sample.

6.3 DETECTORS

Three waterproof detector assemblies were constructed (Figures 24 to 27). These were made of stainless steel (CRES SHT or CRES Tube type 321) to accommodate one of the three sizes of detector used.

The following detectors were used.

1. Sodium Iodide (Thallium activated) in sizes:

1 3/4 inch diameter by 2 inches long

3 inch diameter by 3 inches long

5 inch diameter by 4 inches long

each with aluminum oxide reflective coating.

2. Pilot "B" Plastic Scintillators (Ref. 23) in sizes:

3 inch diameter by 3 inches long

5 inch diameter by 4 inches long

each with aluminum foil reflective coating, loosely coupled to the scintillator.

3. Dr. Miranda's Liquid Scintillant* (Ref. 24) in containers of sizes:

3 inch diameter by 3 inches long

5 inch diameter by 4 inches long

each with the inside of the container coated with vaporized aluminum and containing a quartz end window.

6.4 PULSED HEIGHT COMPARISON OF REFLECTIVE COATINGS

A number of identical plexiglass cyclinders were constructed (3 inches long, 3 inches in diameter, 1/8 inch thick) in order to determine the reflective coating that gives optimum light collection from the

* Dow Corning 555 Fluid plus 100 grams of naphthalene and 4 grams of 2,5 - diphenyloxazole per liter of fluid.

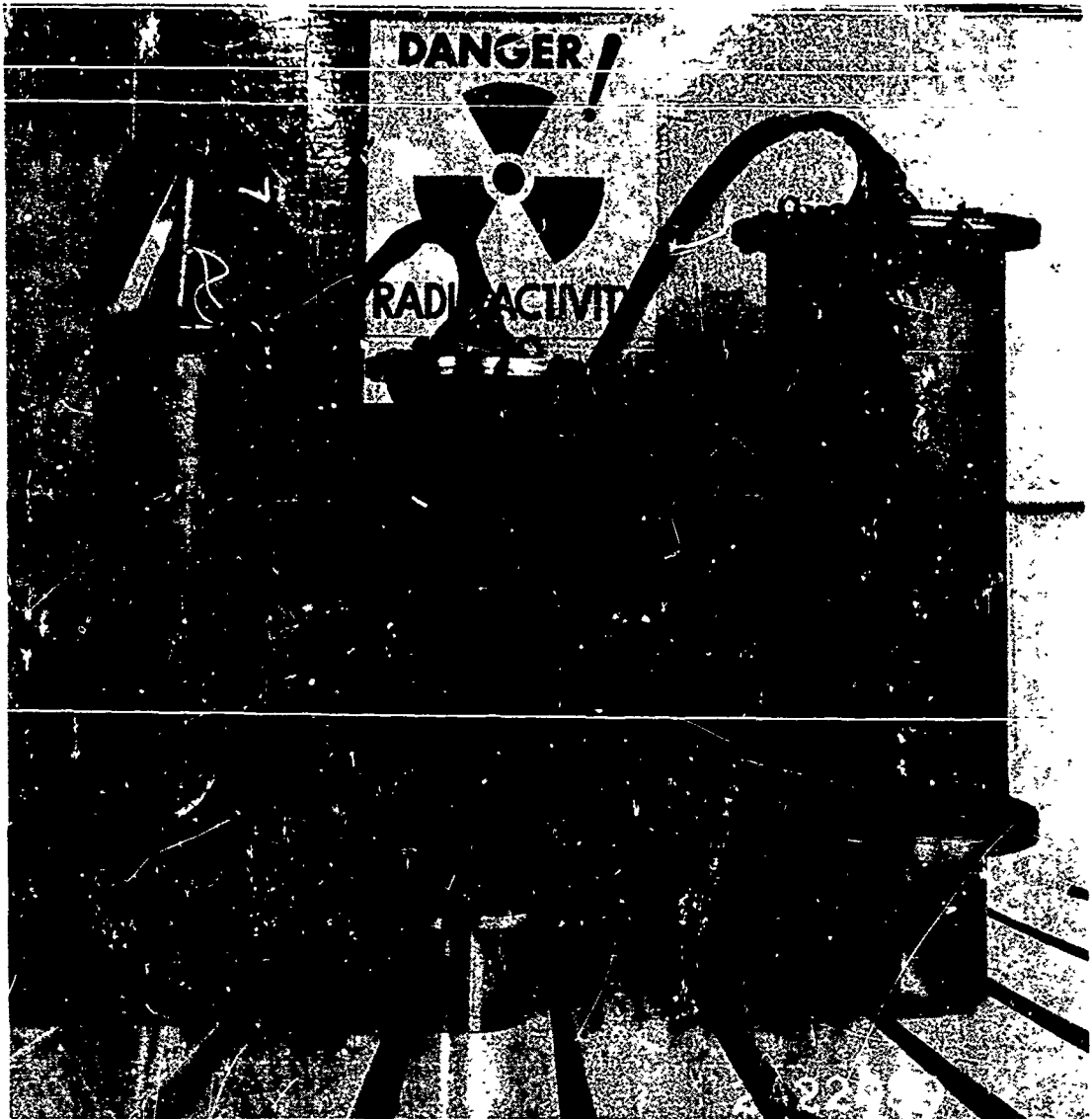


Figure 24. Waterproof Detector Assemblies for Use in the Calibration Tank. The probes, from left to right, would accomodate $1\frac{3}{4}$ -inch by 2-inch, 3-inch by 3-inch, and 5-inch by 4-inch scintillation detectors. NaI(Tl) crystals of all three sizes were tested. Pilot "B" plastic and Miranda's liquid scintillant were tested in the two larger sizes.

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Figure 25. Disassembled View of the 2-Inch Probe. In the foreground are the end plate, a stainless steel sleeve, and the cathode follower attached to the photomultiplier socket. Next, from the left, are the photomultiplier, mu-metal shield, and a 2-inch NaI(Tl) crystal. At the rear is the stainless steel probe housing.

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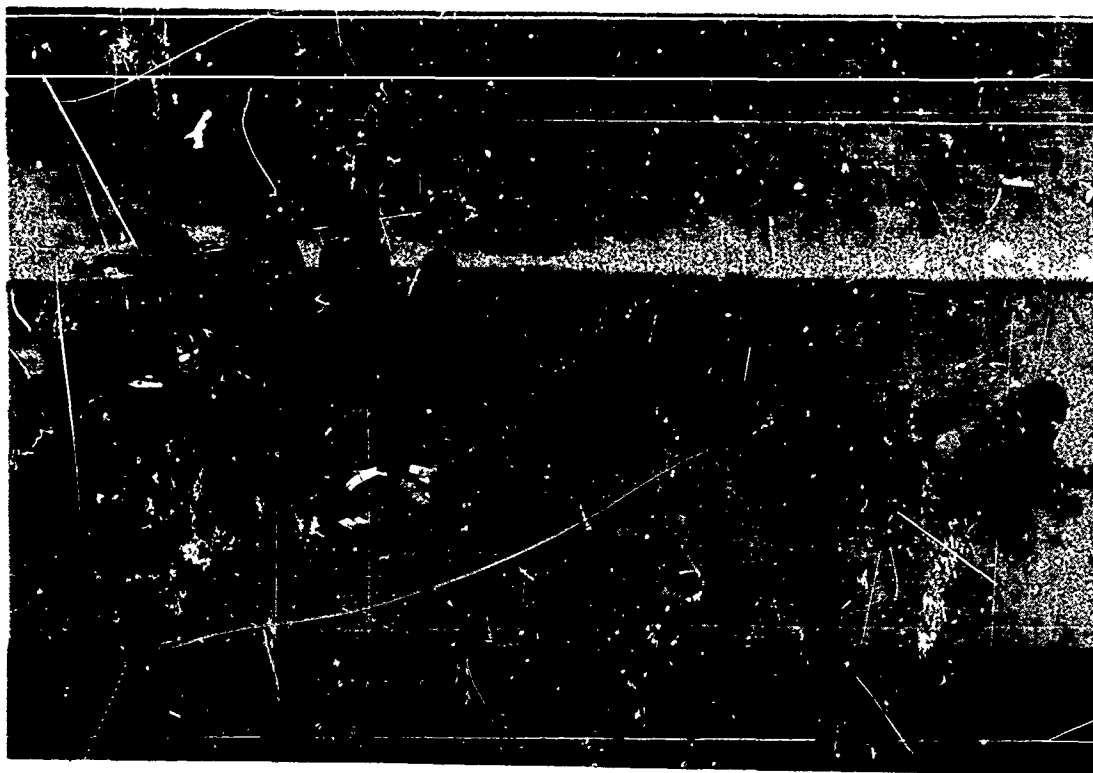


Figure 26. Disassembled View of the 3-Inch Probe. Arranged in clockwise order are the mu metal shield, photomultiplier standing on the plastic phosphor container, sodium iodide crystal container, integral sodium iodide crystal - photomultiplier - shield assembly, probe housing, cathode follower, sleeve, and end plate. In the left foreground are a container and an adapter ring for the liquid scintillant.

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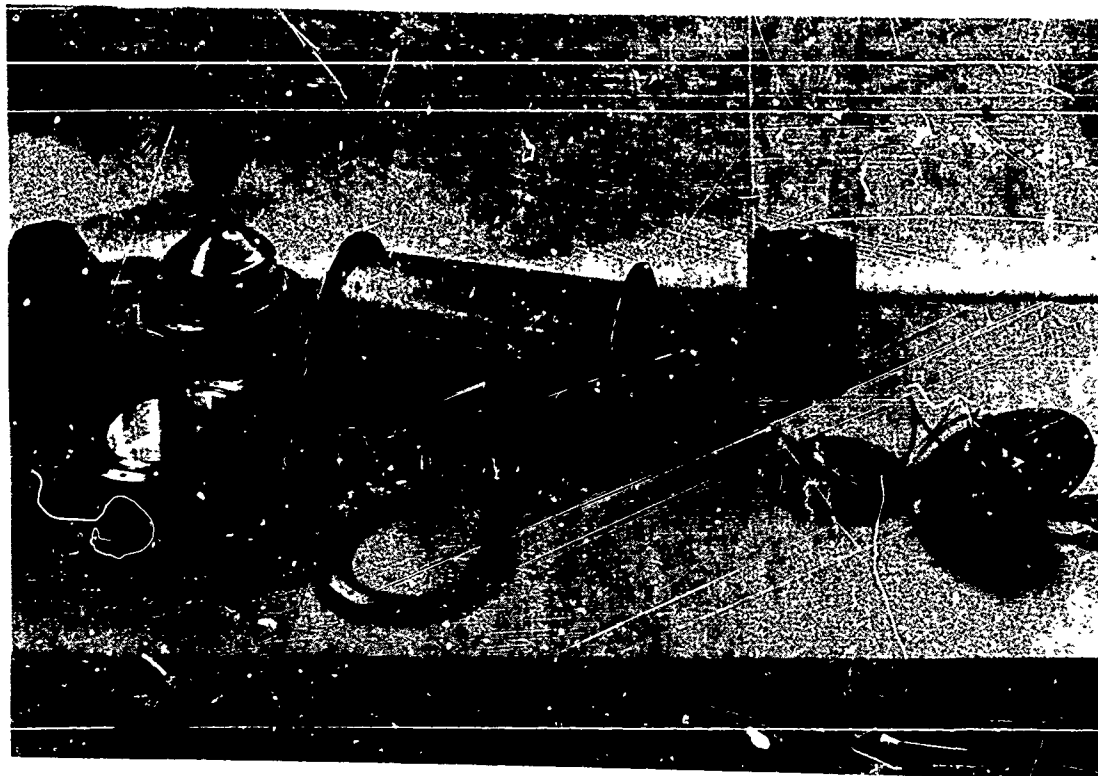


Figure 27. Disassembled View of the 5-inch Probe. A Pilot "B" plastic phosphor, 5 inches in diameter by 4 inches thick is in the container in the left foreground, next to an adapter ring. Behind them are the mu metal photomultiplier shield, 5-inch photomultiplier with the 5-inch by 4-inch NaI(Tl) crystal in its container, the probe housing, cathode follower, a mounting ring, and the end plate. A sleeve is behind the cathode follower.

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liquid scintillant developed by Dr. Henry Miranda of Hudson Laboratory. The emission and absorption spectra of this scintillant are not yet known.

A three-inch DuMont phototube (Type 6363) was fitted with a container which allowed the different cylinders to be easily changed. The cylinders were optically coupled to the phototube with Dow-Corning 200 fluid of relatively low viscosity (1000 centistokes).

The differential pulse-height spectrum was run for each cylinder. The phototube high voltage, 1200 volts, was manually disconnected to change cylinders. A window width of two volts was used (6 mv phototube pulse).

To relate the pulse height of the scintillations to the recorded amplifier output, pulses from the Precision Pulser were introduced at the input of the amplifier, and the pulse height axis was calibrated in terms of cathode follower output voltage; or since the cathode follower has a gain of approximately 0.2, in terms of the phototube output pulse height.

The various coatings used for this experiment and the method of preparation of the various surfaces are listed in Table 7.

The results are shown graphically in Figure 28, showing the counting rate in a narrow "window" ($\Delta V \approx 6$ mv) versus phototube output pulse in millivolts.

The two extreme cases are silver or aluminum evaporated on a smooth surface and silver on a rough surface. The silver on smooth surface gave a mirror-like finish, while the silver on the rough surface

TABLE 7. Preparation of Reflective Coatings.

a. Silver on Smooth Surface (a)

A thin silver film was evaporated on a polished cylinder in a vacuum chamber.

b. Aluminum Foil - Smooth

Aluminum foil, made by Foilrapp Inc., Seattle, was wrapped around the cylinder and secured with a rubber band.

c. Alpha-Alumina - Painted

Alpha-alumina, Type A-5175, Lot No. P-#316, made by Linde Air Products Company, New York, was mixed with Plastic Cement #3, made by Universal Plastics Company, Seattle, and brush-painted on the cylinder. Eight coats were applied.

d. Tygon Paint - Sprayed

White Tygon Paint, TP-61, made by U.S. Stoneware, Akron, Ohio, was sprayed on; approximately 5 coats.

e. Aluminum Foil - Rough

Aluminum foil (same as above) was roughened with alpha-alumina to produce a dull finish, which was wrapped loosely around the cylinder.

f. Magnesium Oxide - Sprayed

Magnesium oxide, "Bakers Analyzed" Reagent grade, made by I. T. Baker Chemical Company, Phillipsbury, New York, was mixed in Glidden's Acrylic Plastic, 170-C-1, clear, and approximately eight coats were sprayed on.

g. Alpha-Alumina - Sprayed

Alpha-alumina, Type A-5175, Lot No. P-#316, made by Linde Air Products Company, New York, was mixed in acrylic plastic, 170-C-1, clear, made by Glidden Co., Cleveland, Ohio. Approximately 15 coats were sprayed on on cylinder.

h. Aluminum on Rough Surface (a)

A thin aluminum film was evaporated on a sand-blasted cylinder in a vacuum chamber.

i. Silver on Rough Surface (a)

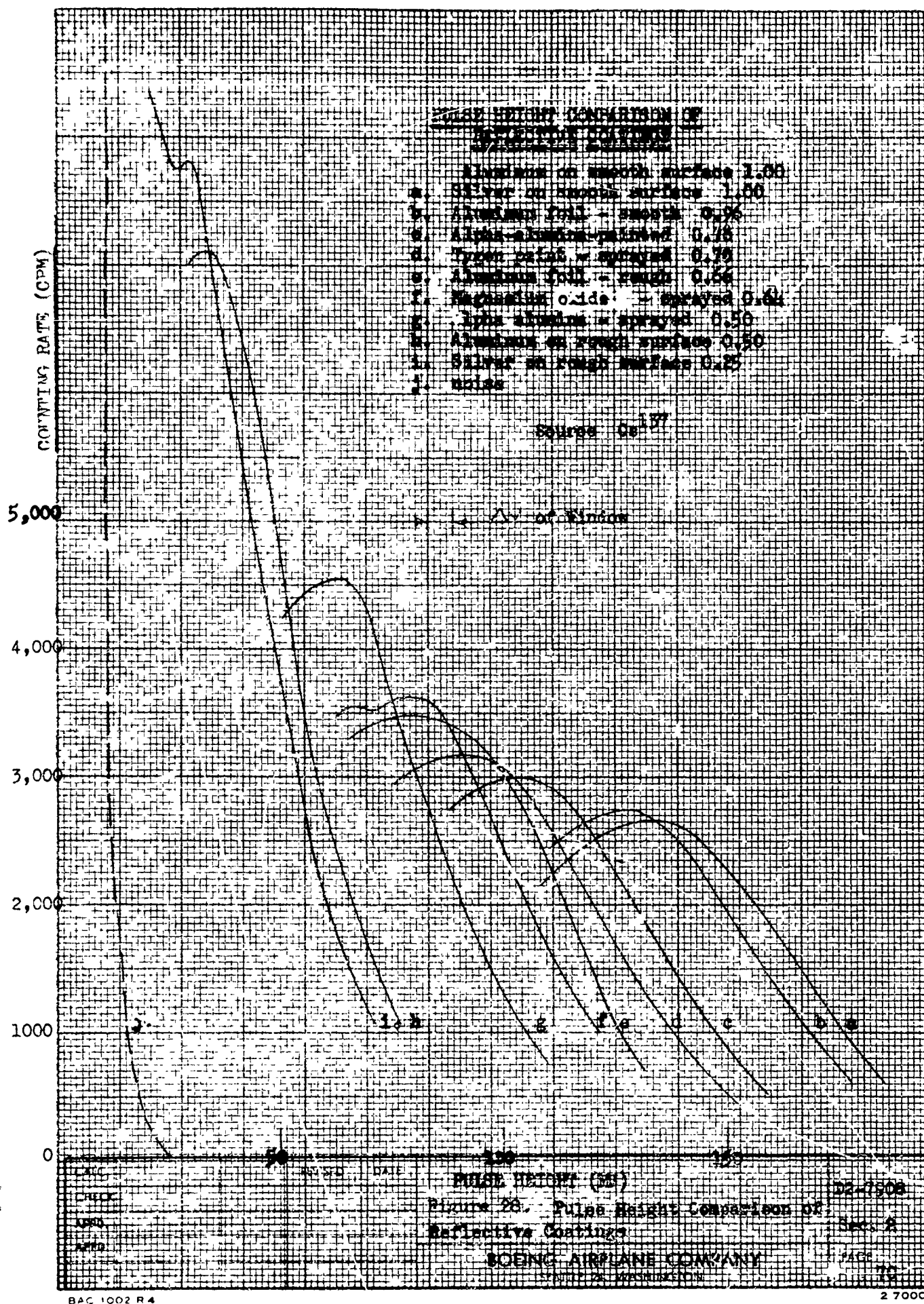
A thin film of silver was evaporated on a sand-blasted cylinder in a vacuum chamber.

NOTE: (a) Prepared by BAC Optics Laboratory

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looked dark gray to the eye. There is a wide difference between alpha-alumina sprayed on and painted on. In the first case, although 15 coats were applied, the coating was thin and appeared somewhat opaque, while the painted coat was much thicker, having a white chalky appearance. Apparently the coating with the greatest reflectivity performs best for this scintillant. Next came the diffuse reflectors, where those with the "whitest" appearance perform best, up to about 75 percent of the "mirror" finish. It should be noted that the silver and aluminum coatings evaporated on a smooth surface gave essentially identical results.

The fact that the liquid scintillant used in this experiment was bubbled with dry nitrogen may have produced misleading results because: although larger pulses can be obtained from a given scintillant by driving off the dissolved oxygen, the behavior of the scintillant is not as reproducible unless care is taken to handle it under a nitrogen atmosphere, in filling the containers, etc. Because of this, many experiments of a comparative nature are performed using air-saturated scintillant (i.e., no steps are taken to remove oxygen). Although the pulses are smaller, the performance is said to be more reproducible.

6.5 TANK

The tank (Figure 29) was cylindrical, 85 inches high with a mean diameter of 92-5/8 inches. It was constructed of Douglas Fir to minimize scattering, and the inside was lined with Fiberglass, to permit decontamination. The water level was maintained at 80 inches from the bottom, giving a usable volume of 8.834×10^6 cubic centimeters. The

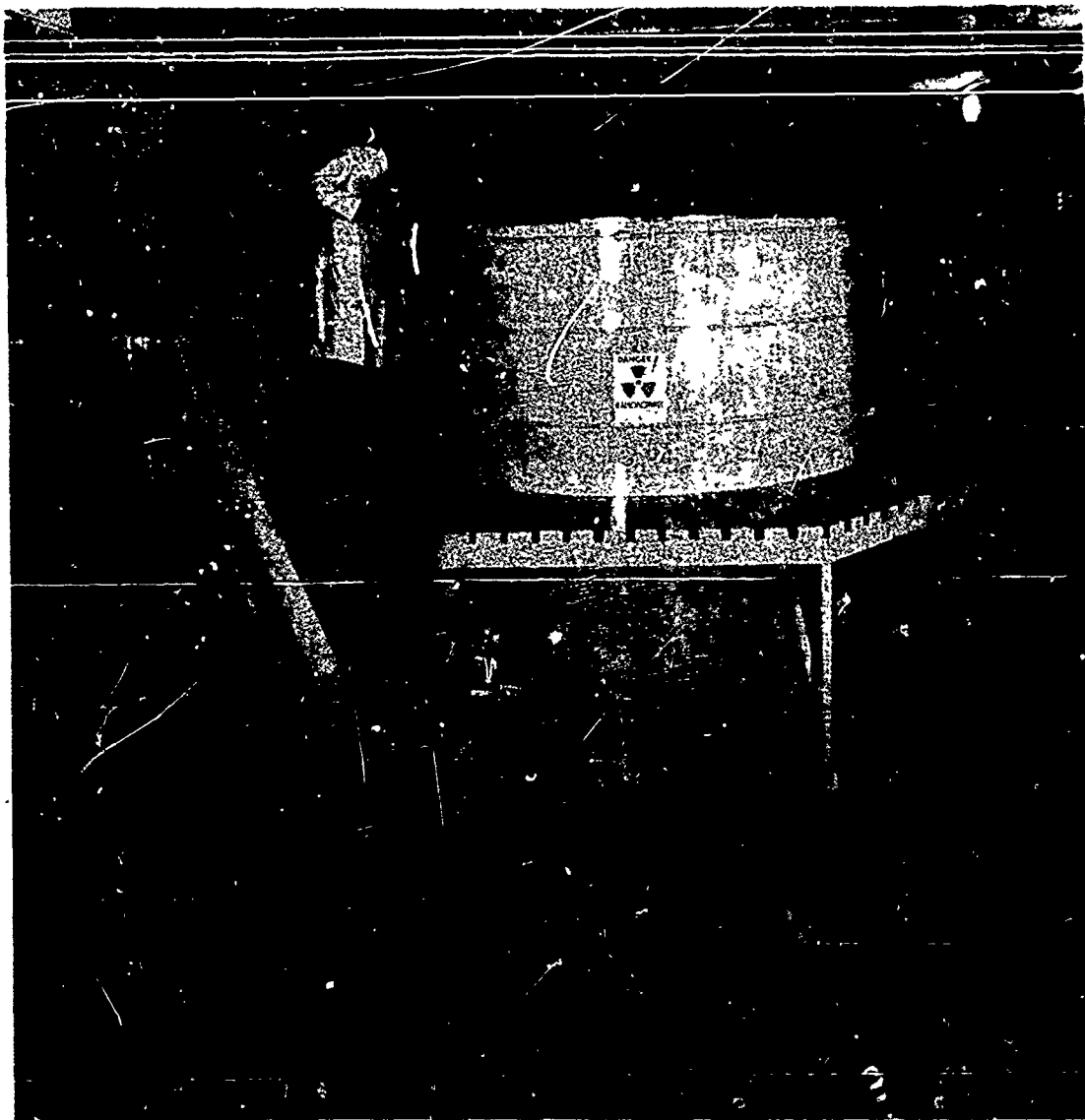


Figure 29. 2500 Gallon Tank for Study of Volume Distributed Gamma Sources. Constructed of Douglas fir and lined with fiberglass, the tank is approximately 8 feet high by 8 feet in diameter and contained 2300 gallons of the test solutions. The photopeak count rate with Na^{24} in the tank (gamma energies of 1.37 and 2.75 Mev) is only about 2% less than the count rate that would be obtained with the detector in a solution of the same composition but of infinite volume.

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waterproof probe was suspended in the tank, with the center of the scintillator 40 inches from the bottom of the tank (Figure 30).

6.6 INSTRUMENTATION

All spectra were taken with a Radiation Counter Laboratories, Inc. Model 20611, 256-channel pulse height analyzer (Figure 31). It provided a cathode-ray tube presentation of the accumulating spectrum for monitoring purposes, a strip chart recorder, and a digital printer. It can store a maximum of $2^{16} - 1$ or 65,535 counts in each channel, and can handle approximately 10^5 counts per second without data distortion.

The linearity of the analyzer is ± 0.5 percent of full scale, except for the first 5 channels.

The "dead time" is somewhat less than $(20 + 0.5 N)$ microseconds, where N is the number of channels in which a pulse is stored. However, the analyzer has a "live-time" indicator, which will count time only when the input gating circuits are accepting pulses, making a dead-time correction of the data unnecessary.

6.7 ENERGY CALIBRATION

The detectors were calibrated for energy response before each measurement in the tank, using at least three test sources with convenient energies (mercury-203 = 0.279 Mev, cesium-137 = 0.662 Mev and yttrium-88 = 0.91 Mev and 1.85 Mev). Since separate energy calibrations were run for each measurement, no attempt was made to keep the same amplifier gain settings and photomultiplier high voltage.

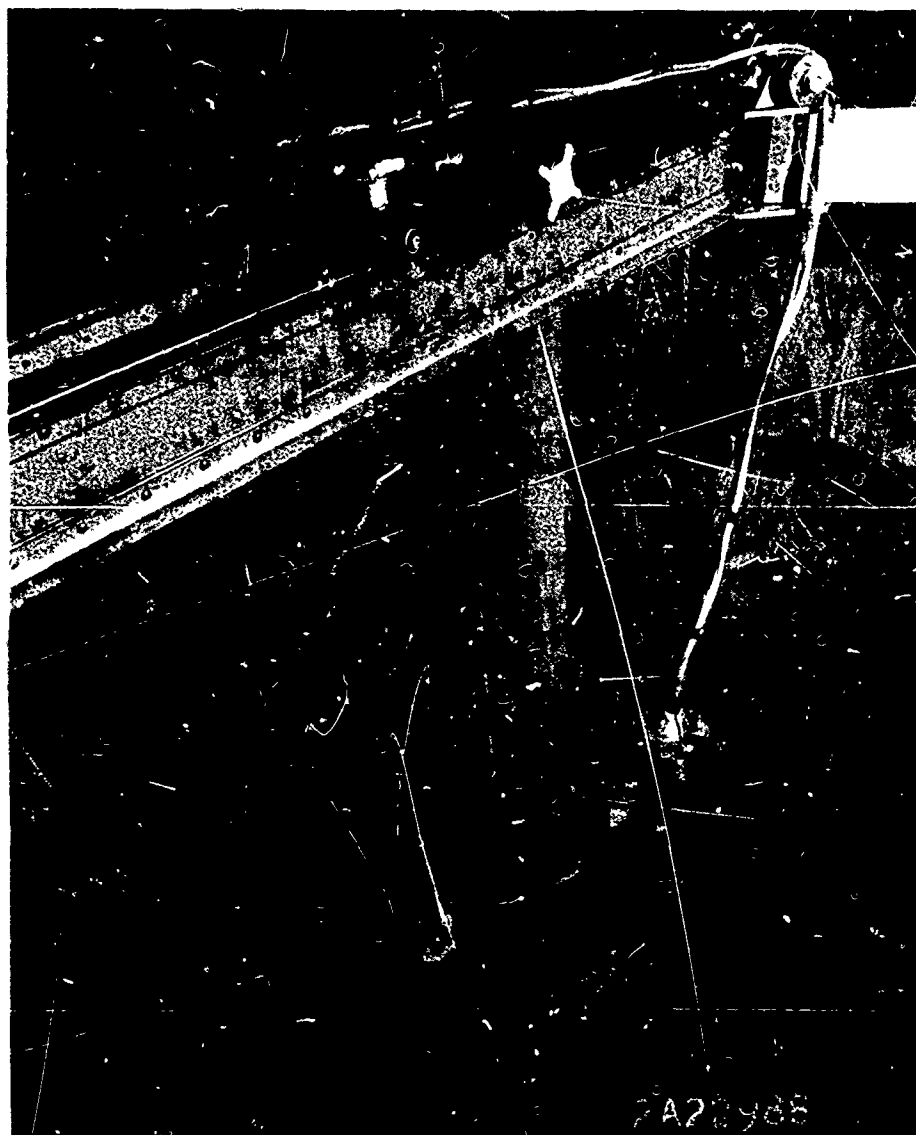


Figure 30. Internal View of the Tank with the 3-Inch Probe in Central Position. A discrete source, at the end of the vertical rod, can be moved in relation to the detector for study of attenuation and scattering of gamma rays in water (unpublished).

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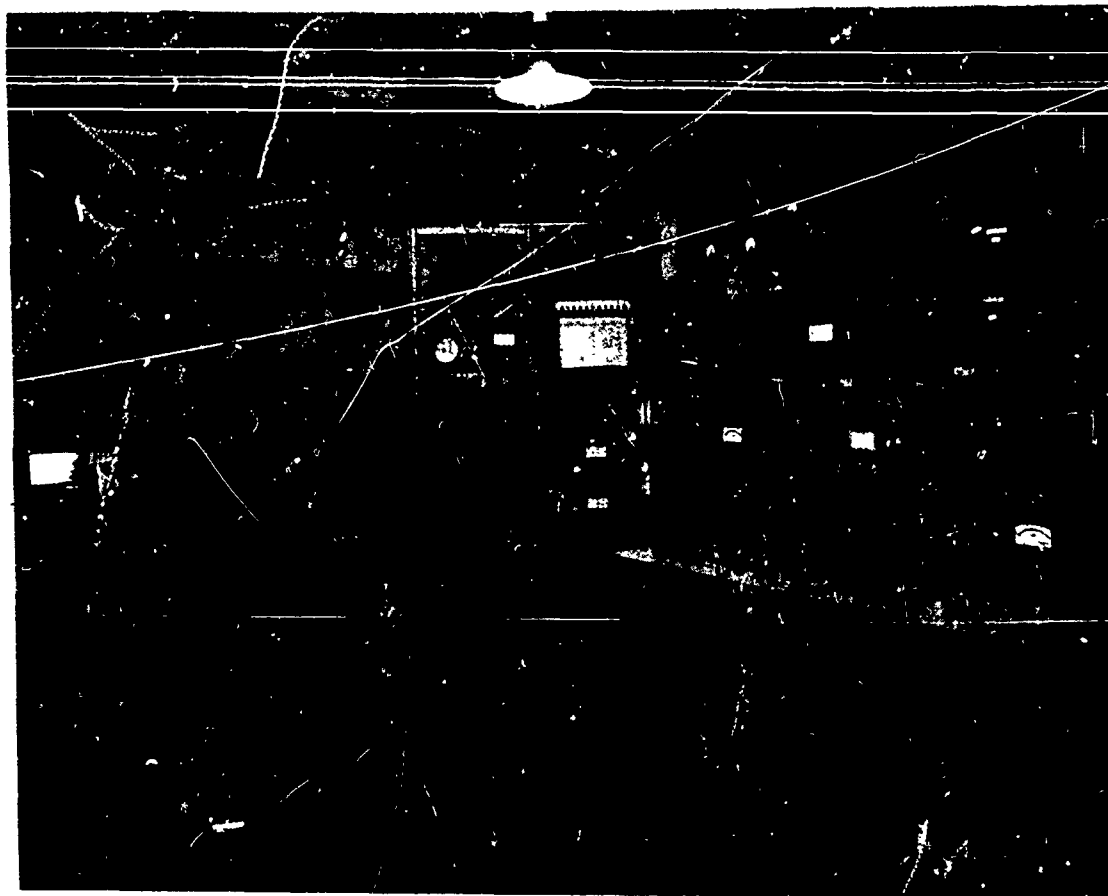


Figure 31. View of the Counting Room Showing the Shield for Low Level Counting, 256 Channel Analyser, and Electronic Equipment Rack with Various Amplifiers, Analysers, Scalers, and Power Supplies.

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6.8 ISOTOPE MIXING

When using chlorine-38 or sodium-24, the isotope was dissolved in 300 ml of water. From this, a 10 ml sample was placed in a 25 ml volumetric flask, which was placed in a 50 ml beaker. The beaker was then placed directly on top of the 3-inch well type crystal to determine its specific activity as described in the next part.

The remaining 2990 ml were poured into the tank and mixed. The tank bottom was equipped with a compressed air line with a number of small air holes to permit violent bubbling of the contents.

It is estimated that uniform mixing occurred within 1 to 2 minutes of bubbling. To verify this, after an experiment using the muriate of potash the tank was allowed to mix for approximately an hour, and the experiment was repeated without significant change in counting rates.

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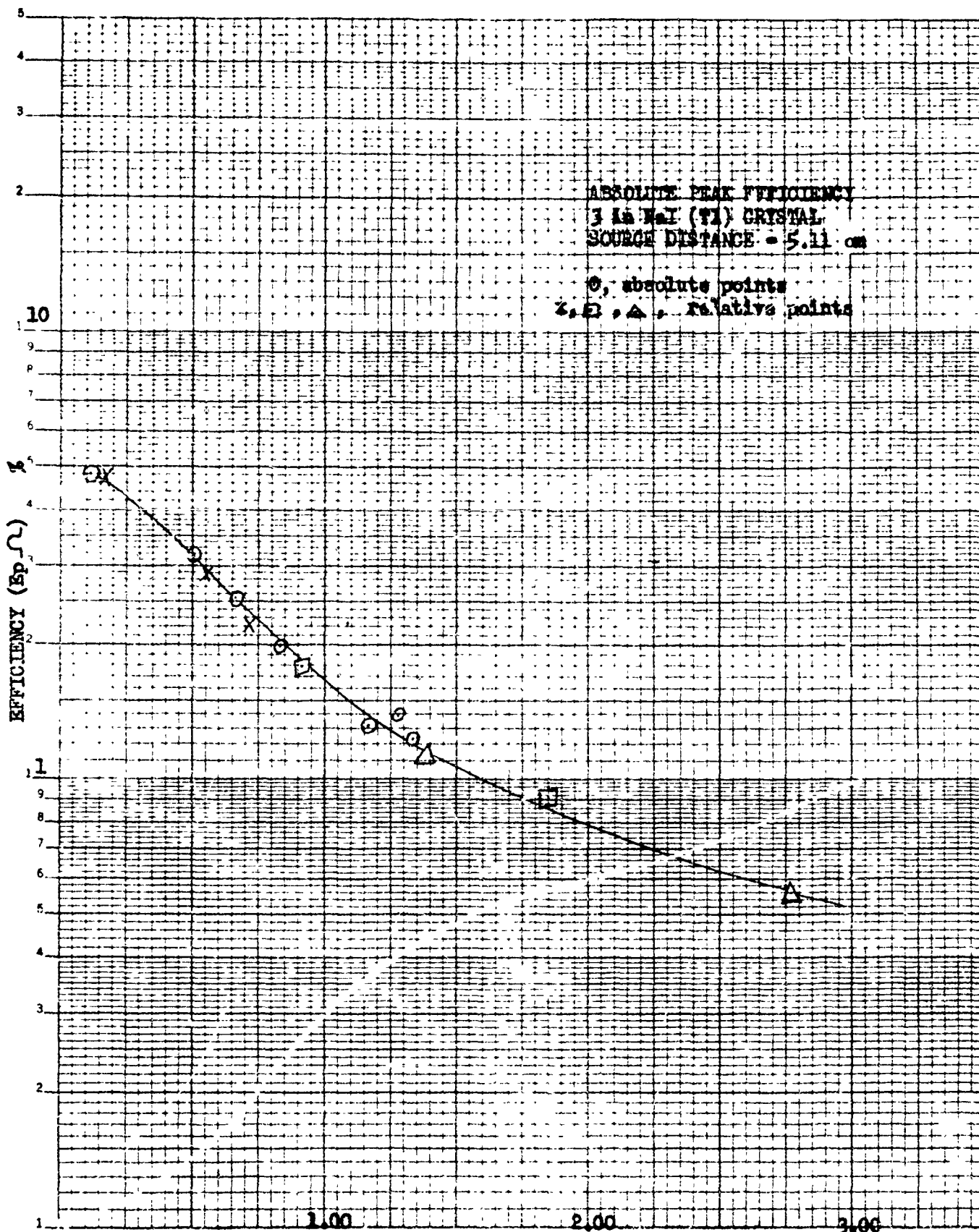
7. SOURCE STRENGTH DETERMINATIONS

The source strengths of samples introduced into the tank were measured by placing a 10 ml sample on top of the three-inch well type crystal. The 10 ml sample was contained in a 25 ml volumetric flask (which in turn was placed in a 250 ml beaker to avoid contaminating the crystal in case of spills). This geometry was used for sodium-24 and chlorine-38 strength determinations.

To relate the photopeak areas of the above measurements to the source strength, the 3 by 3 inch NaI crystal was calibrated using a set of standard sources (Ref. 25) and a source to detector distance of 5.11 cm. The absolute photopeak efficiency ($\epsilon_{p\Omega}$) of the crystal was determined as a function of gamma energy with the following standard sources:

<u>Isotope</u>	<u>Energy</u>	<u>Source Strength</u>
Cobalt-57	0.123 Mev	1480 γ 's/sec
Sodium-22	0.511 Mev	2060 γ 's/sec
Cesium-137	0.662 Mev	3030 γ 's/sec
Manganese-54	0.840 Mev	3140 γ 's/sec
Cobalt-60	1.17 Mev	2240 γ 's/sec
Sodium-22	1.28 Mev	1150 γ 's/sec
Cobalt-60	1.33 Mev	2240 γ 's/sec

In addition to these "absolute" points, spectra of yttrium-88 and sodium-24 sources were taken also 5.11 cm. distance, to give two more pairs of "relative" efficiency points, which were then fitted to the absolute efficiency curve (Figure 32).



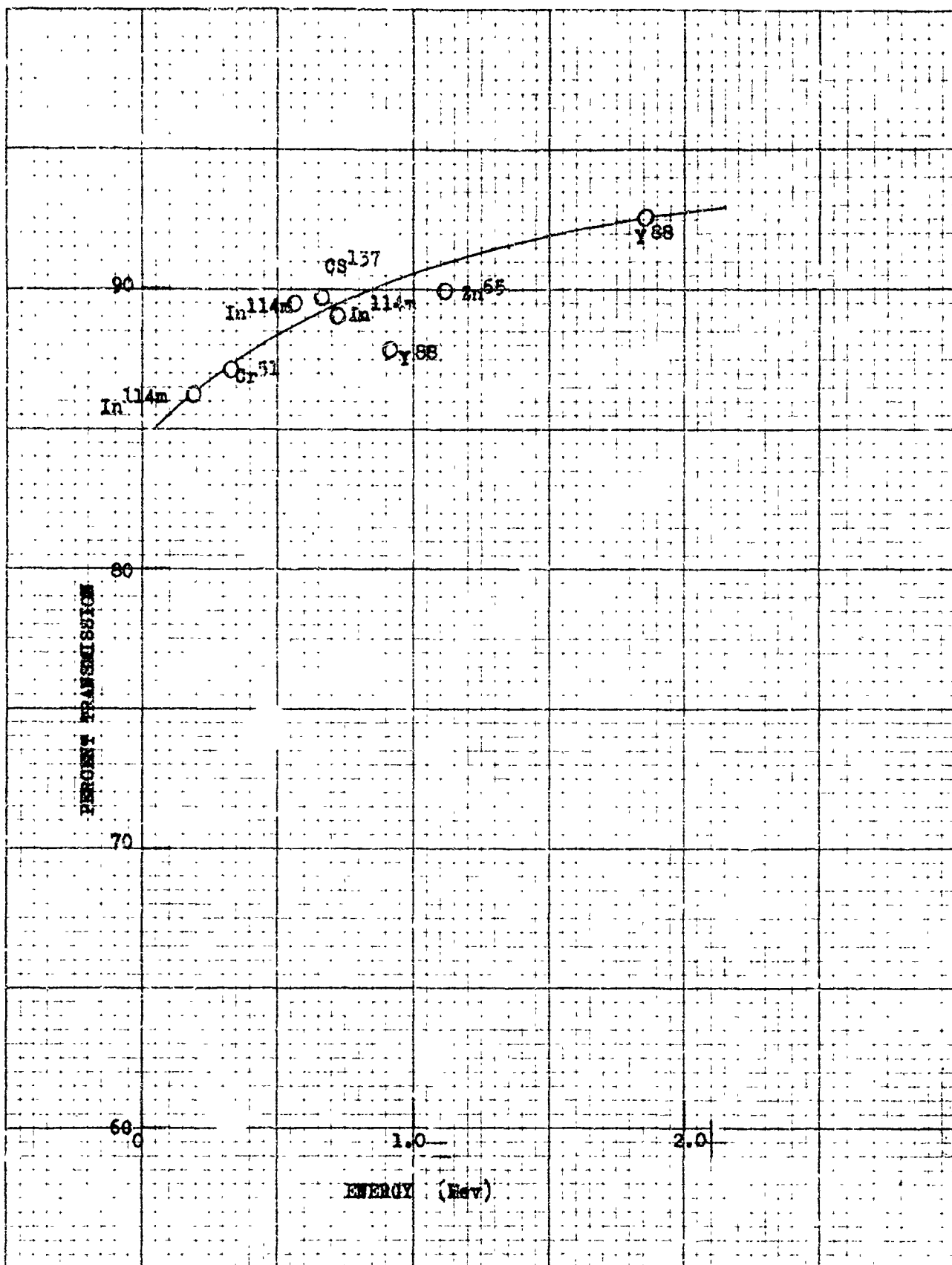
CALC		REVISED	DATE	ENERGY (MeV)	
CHECK				Figure 32. Energy Calibration of 3-inch NaI(Tl) Crystal	D2-7908
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The effect of the (0.018) stainless steel cover of the 3 x 3 inch crystal, the 1/16 inch layer of aluminum oxide, and the 1/2 inch diameter, 7/16 inch long lucite rod (part of the standard source) on the photopeak area was measured using sources evaporated on mylar film at a distance of 5.11 cm with and without a dummy absorber and plastic rod. The reduction in photopeak area varied from 14 percent at 190 Kev to 8.5 percent at 1.85 Mev. The curve showing percent transmission vs. energy is shown in Figure 33.

The photopeak areas were further corrected for coincidence summing, since for sources which emit two or more photons, certain number of counts which normally fall in one photopeak are lost because they are detected simultaneously with a photon of another gamma energy. This "sum" peak also has an accompanying Compton distribution, i.e., peak counts of energy 1 in coincidence with Compton counts of energy 2, etc.

The correction term is obtained as follows:

Let	$P(\gamma_i)$	be the probability of detecting a gamma ray of energy (i) in the photopeak,
	S	be the source strength in dist/sec.,
	$[E_p]_{\gamma_i}$	be the photopeak efficiency of the detector for γ_i ,
	$[E_t]_{\gamma_i}$	be the total efficiency for the detection of γ_i ,
	Ω	be the solid angle subtended by the detector,
	K_i	be the fraction of gamma rays γ_i per disintegration,
	α_{ij}	be the number of gamma rays γ_j in coincidence with γ_i ,



CAIC		REVISED	DATE	<p>Figure 33. Percent Transmission with Absorber, 0.020 inch Stainless Steel and 1/16 inch Magnesium Oxide.</p> <p>BOEING AIRPLANE COMPANY</p>	D 2-7908
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A_2 be the fraction of γ_2 transmitted through any absorbing material present.

Then:

$$P[\gamma_1] = SA_1 K_1 [\epsilon_p \Omega]_{\gamma_1} - SA_1 K_1 [\epsilon_p \Omega]_{\gamma_1} A_2 \alpha_{12} [\epsilon_1 \Omega]_{\gamma_2}$$

That is, the number of counts in peak (γ_1) will be the product of the source strength and the photopeak efficiency, minus the number which are lost due to summing. The number lost is, in turn, equal to the source strength times the probability of detecting a count in the peak times the probability of simultaneously detecting a photon of energy γ_2 (either peak or Compton). Hence the total efficiency for γ_2 is used. The total efficiency was obtained from R. L. Heath (Ref. 26).

The source strength is now given by:

$$S = \frac{P[\gamma_1]}{K_1 A_1 [\epsilon_p \Omega]_{\gamma_1} [1 - A_2 \alpha_{12} (\epsilon_T \Omega)_{\gamma_2}]}$$

The correction term:

$$[1 - A_2 \alpha_{12} (\epsilon_T \Omega)_{\gamma_2}]$$

was applied to all isotopes which had gamma rays in coincidence. These were cobalt-60, yttrium-88, sodium-24, chlorine-38, and indium-114 metastable. The data are plotted in Figure 32.

The absolute efficiency curve for the 3 x 3 inch NaI(Tl) crystal was plotted, for source to detector distance of 5.11 cm and is shown in Figure 32.

The source strength of a "point" source of sodium-24 and chlorine-38 was then determined using the above efficiency curve. The point source was then dissolved in a 10 ml solution, and placed in the standard geometry used for all source strength determinations, giving the photoppeak area in this geometry as a function of source strength determined at 5.11 cm.

The source strength of the potassium-40 sample was determined by chemical analysis of the potassium chloride.

Since the sodium-24 and chlorine-38 samples were decaying while measurements of the source strength was being performed, the actual measured spectrum corresponded to an average source strength in the tank between the times $t = 0$ and $t = t_1$, during which the measurements were made. Since radioactive substances follow the decay law:

$$N = N_0 e^{-\lambda t}$$

the measured activity N_{meas} was:

$$N_{\text{meas}} = \frac{1}{t_1} \int_0^{t_1} N d\tau = \frac{N_0}{\lambda t_1} \left[1 - e^{-\lambda t_1} \right]$$

This decay correction was made and the source strength was determined for $t = 0$.

7.1 TREATMENT OF DATA

The data as presented by the 256-channel analyzer were histograms of the number of counts in the energy intervals defined by the channel boundaries. These data were converted to counting rates $N(\text{chan})$ by dividing by the live time of the analyzer for the particular run. An

energy versus channel number calibration curve was obtained for each run from the spectra of a series of isotopes with gamma rays of known energy. This relationship was linear and described by the equation:

$$E = a(\text{channel number}) + b$$

where a is the channel width and b is the calibration constant

From this information a plot was made of the counting rate per unit energy $\overline{N(E)}$ as a function of the energy. To get $\overline{N(E)}$ from $N(\text{chan})$ divide $N(\text{chan})$ by the parameter a of the energy vs. channel number relationship. This relation may be derived as follows:

$$\overline{N(\text{chan } K)} = \int_{E_{\text{chan } K}}^{E_{\text{chan } (K+1)}} N(E) dE = \overline{N(E)} \int_{E_{\text{chan } K}}^{E_{\text{chan } (K+1)}} dE = \overline{N(E)} \left[\frac{dE}{d(\text{chan})} \right] \int_K^{K+1} d(\text{chan})$$

$$\overline{N(E)} = N(\text{chan}) \frac{d(\text{chan})}{dE} = \frac{N(\text{chan})}{a}$$

where $\overline{N(E)}$ is the average counting rate in the energy interval.

Since the source strengths varied from run to run, the results were normalized to an arbitrary value of 10^{-6} $\mu\text{C/ml}$ for the chlorine-38 and sodium-24 data and the average sea water activity of 3.3×10^{-7} $\mu\text{C/ml}$ for the potassium-40 data in order to be able to compare the results. These are the curves which are plotted as the differential spectra. To obtain the integral spectra the total number of counts above a given energy was determined by summing the differential curves and the results were plotted as functions of the energy.

Just as in the case of the source strength determination, the spectra of sodium-24 and chlorine-38 decay correction had to be made. Since radioactive substances decay as:

$$N = N_0 e^{-\lambda \tau}$$

the average activity in the tank between the times t_1 and t_2 during which the measurements were made was:

$$N = \frac{1}{\Delta \tau} \int_{\tau_1}^{\tau_2} N d\tau = \frac{N_0}{\lambda \Delta \tau} (e^{-\lambda \tau_1} - e^{-\lambda \tau_2}) \quad (U)$$

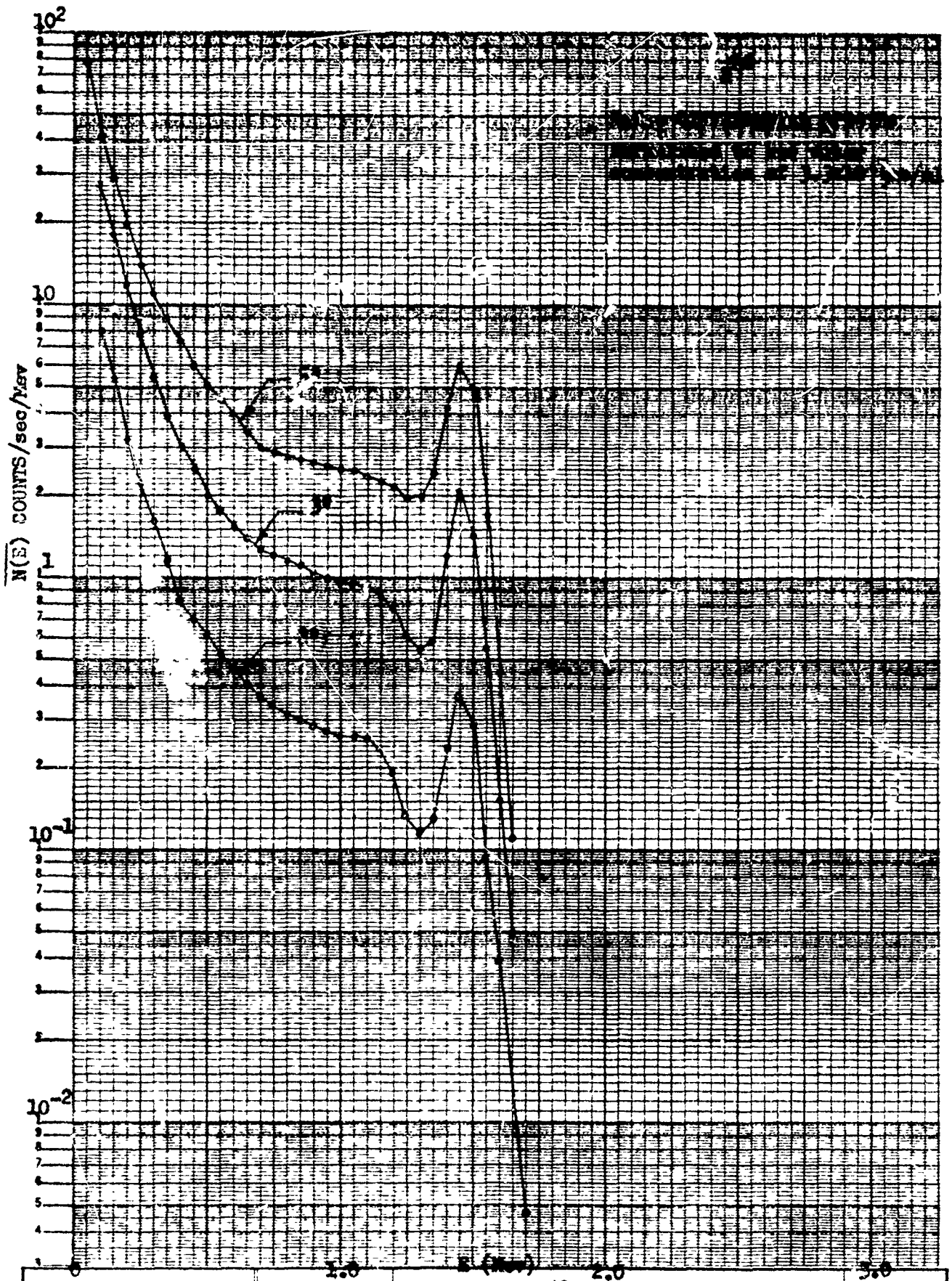
where $t = t_2 - t_1$ is the counting interval. Time $t = 0$ was the time at the beginning of the determination of the source strength for which the initial source strength was calculated. (U)

7.2 SPECTRA

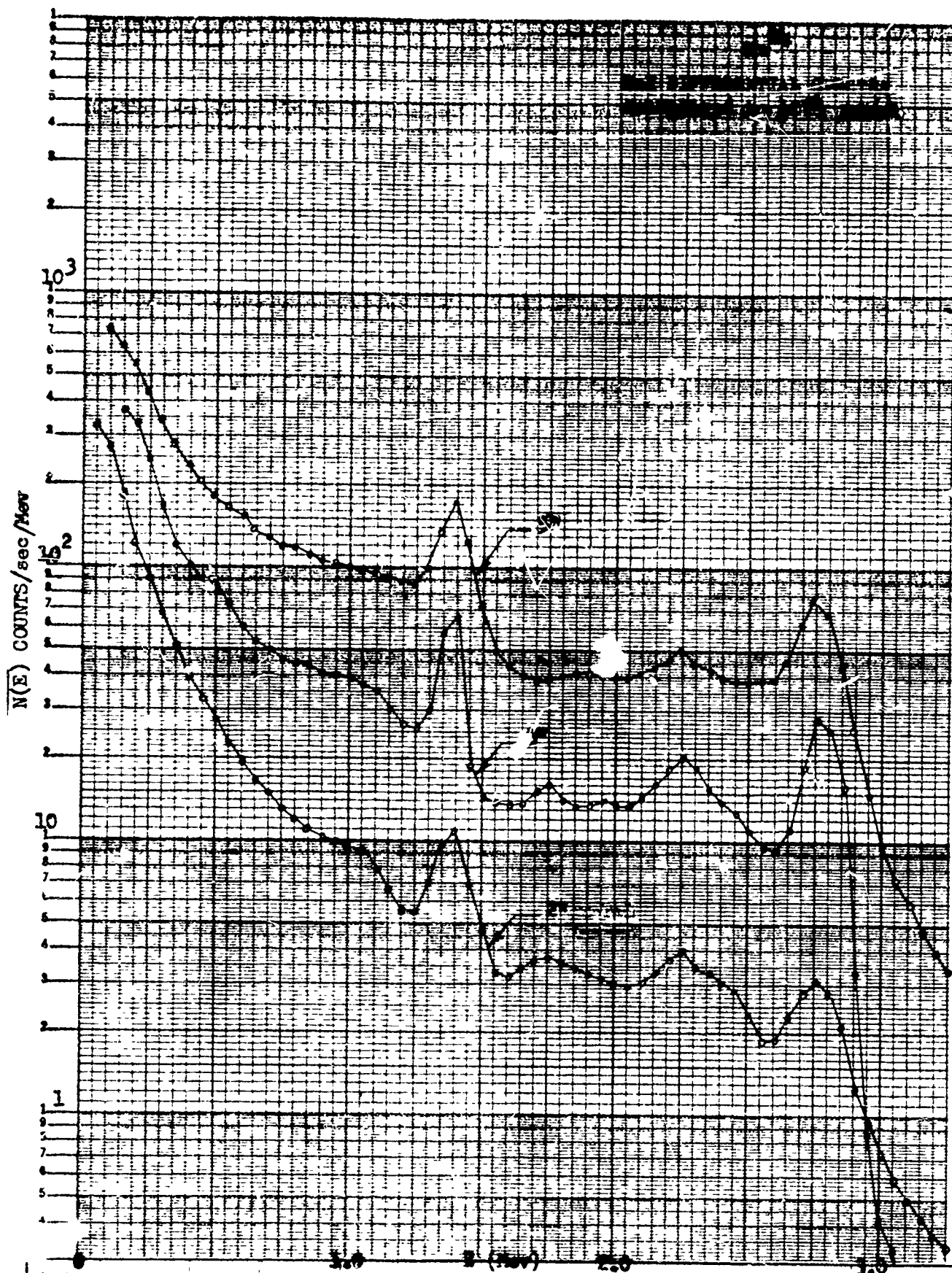
The results of the experiment are summarized in the differential and integral spectra shown in Figures 34 to 45. Spectra were obtained for volume distributed sources of potassium-40, sodium-24 and chlorine-38. The sodium-24 and chlorine-38 sources were normalized to a specific activity of 10^{-6} $\mu\text{c/ml}$ and the potassium-40 was normalized to 3.3×10^{-7} $\mu\text{c/ml}$ (which is the average potassium-40 activity in sea water). (U)

Spectra were obtained for 2, 3 and 5 inch NaI crystals and 3 inch and 5 inch Pilot B plastic phosphors. From these spectra, composite spectra can be obtained which give the spectra which would result from various values of neutron leakage flux and at various times after the passage of the submarine. Different sizes were chosen so as to show the relative effects of varying the dimensions of the detector. (C)

The change in spectral shape with time of the induced activities of sodium-24 and chlorine-38 is shown in Figure 46. The chlorine-38 and sodium-24 spectra are combined at $t = 0$ in the ratio of 7.4 to 1 which (U)



CALC CHECK APR APR	REVISED DATE	Figure 34. K^{40} Differential Spectra for NaI(Tl) Crystals	D2-7908
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Figure 35. Na^{24} Differential Spectra for NaI(Tl) Crystals

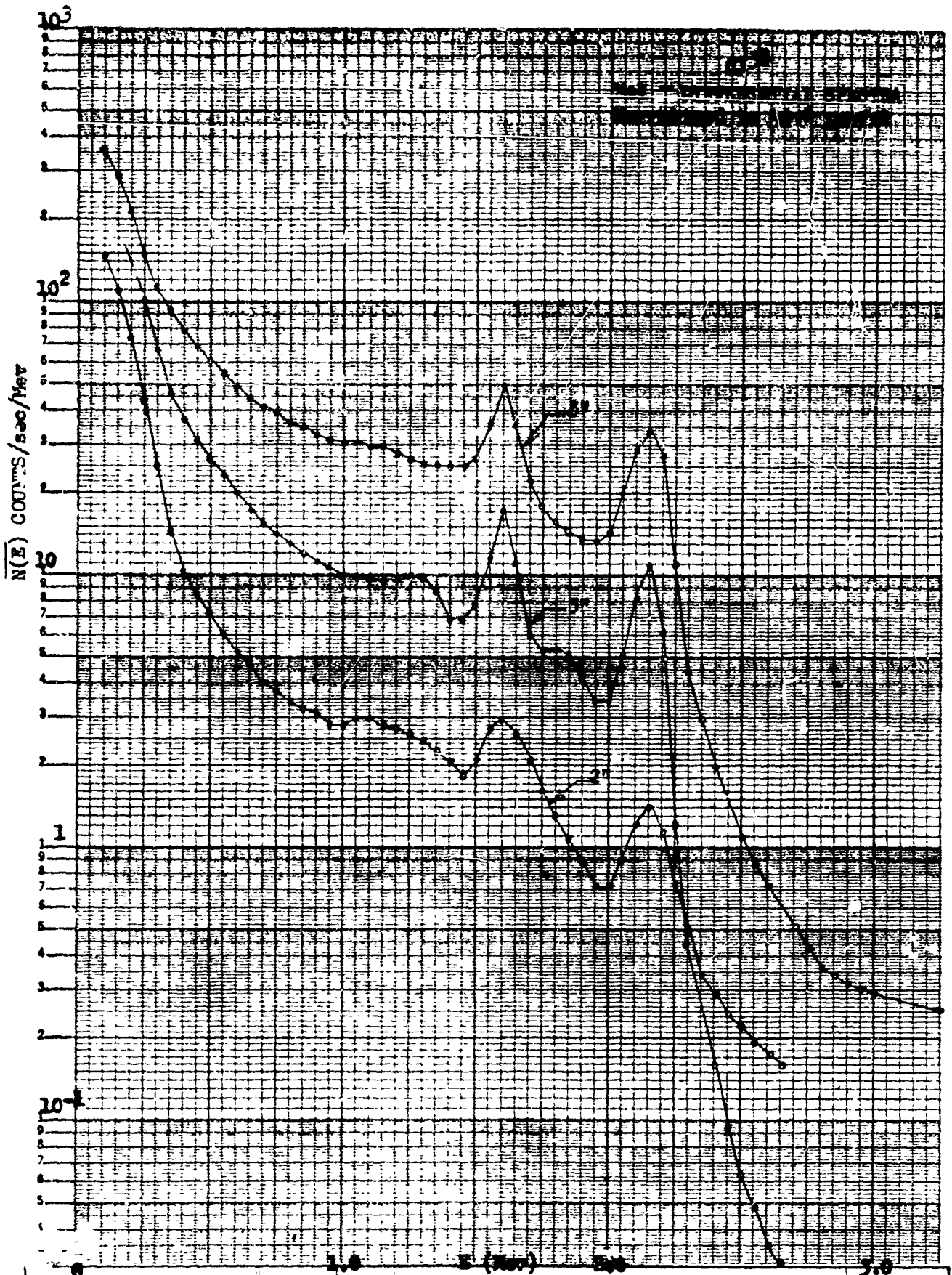
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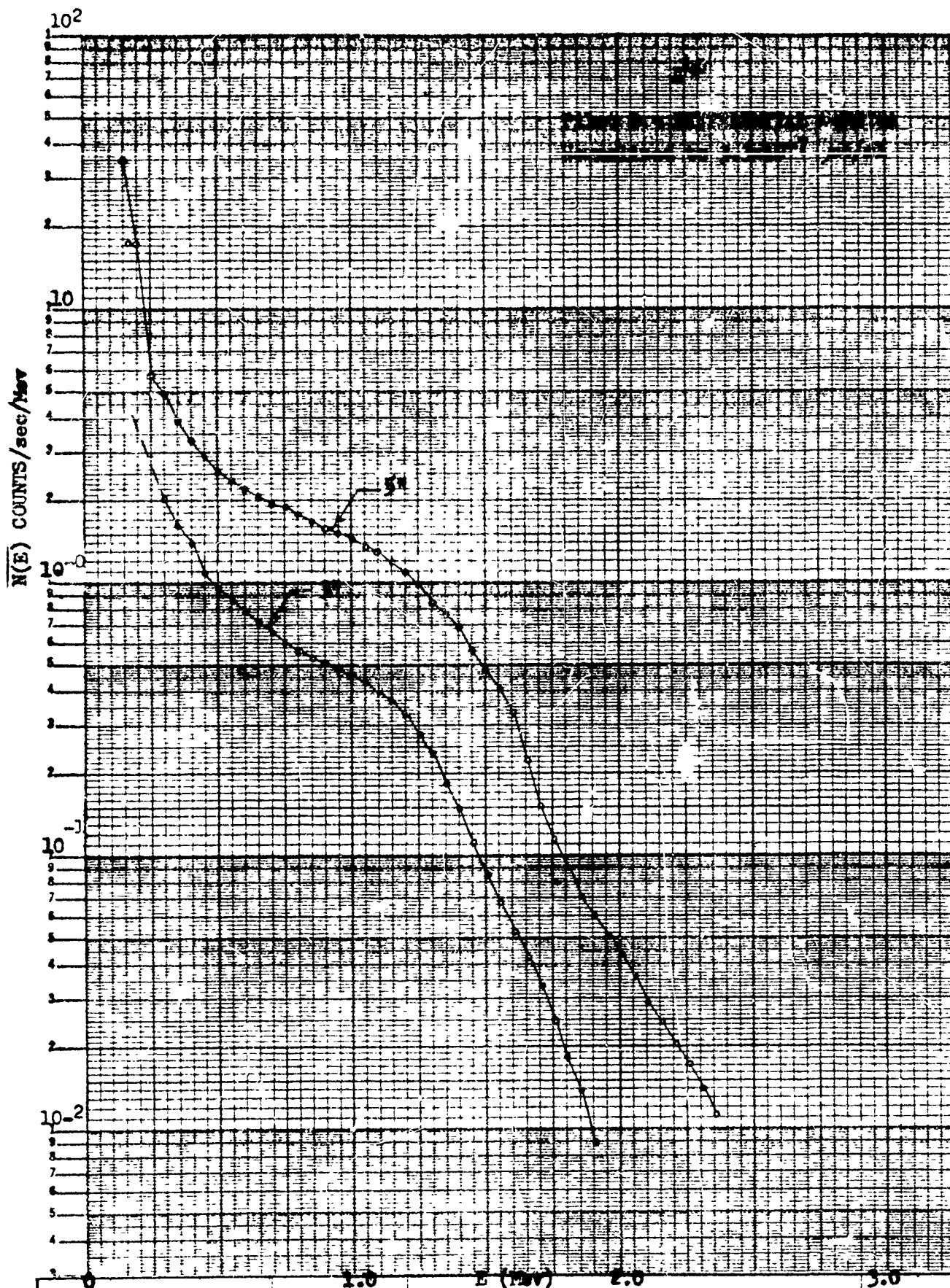
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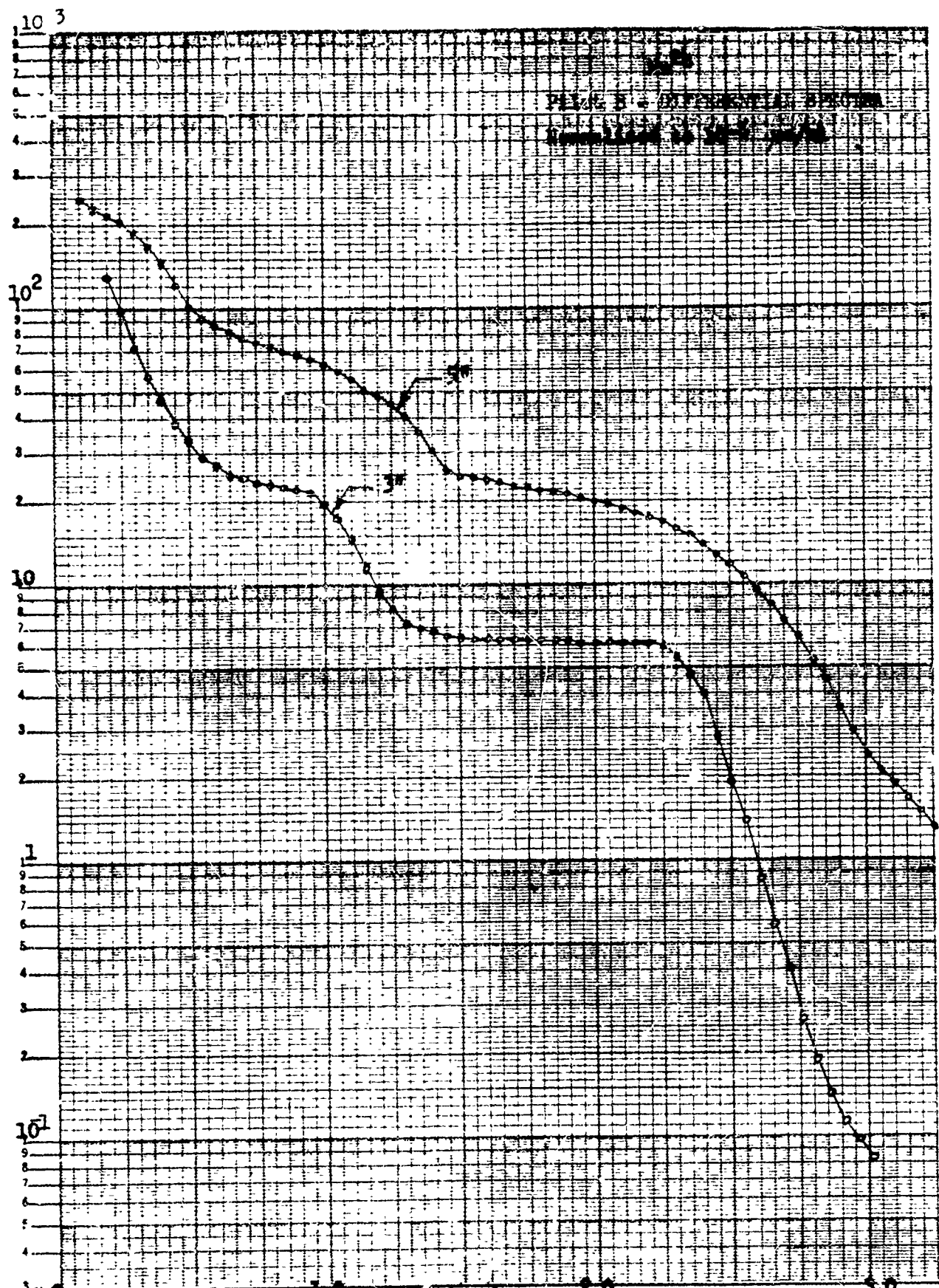


CALC CH CK APR APR	REVISED DATE	Figure 36. Cl^{38} Differential Spectra for NaI(Tl) Crystals	D2-7908
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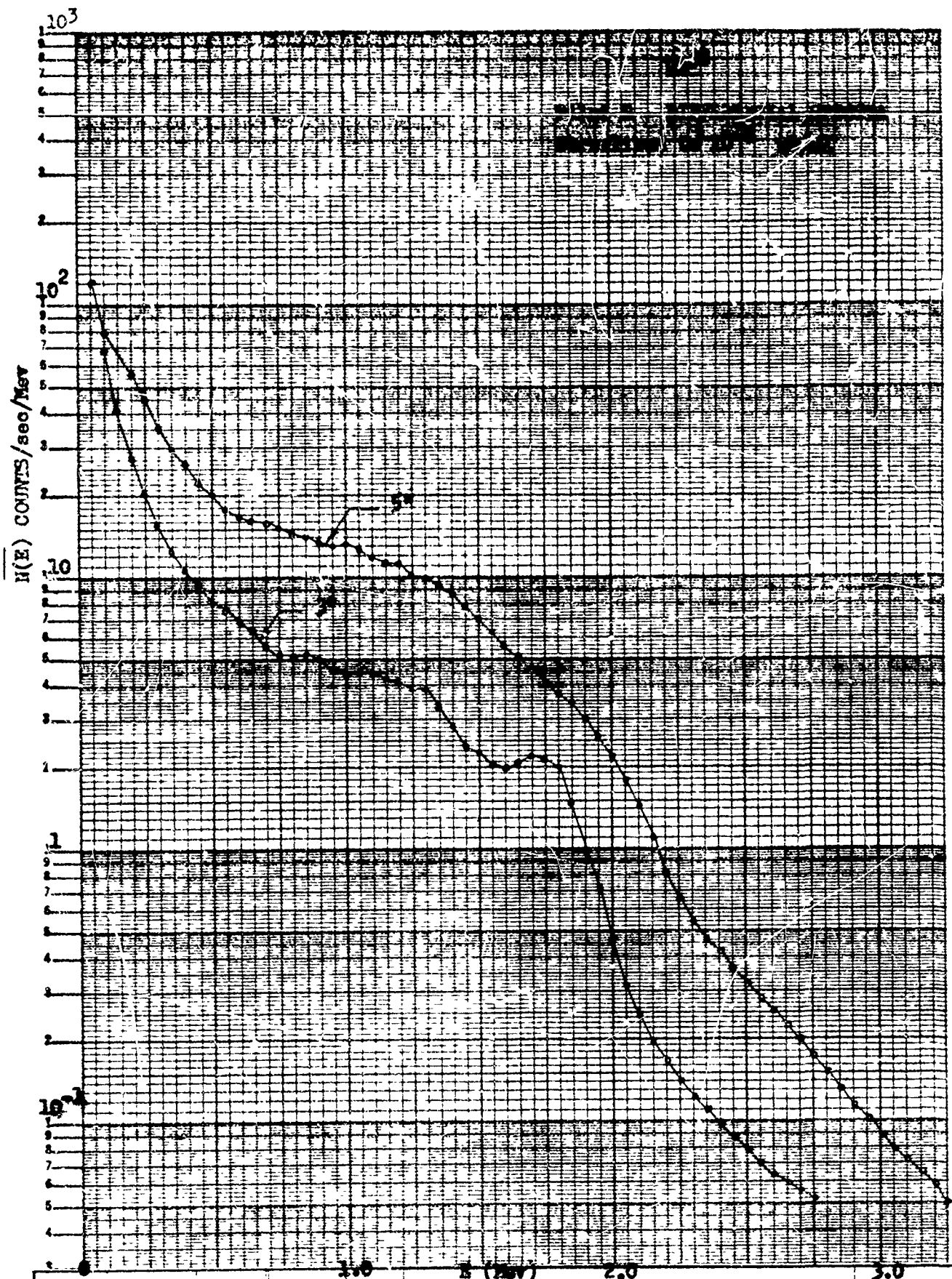


CALC		REVISED	DATE	Figure 37. K^{40} Differential Spectra for Pilot B Plastic Phosphors	D2-7908
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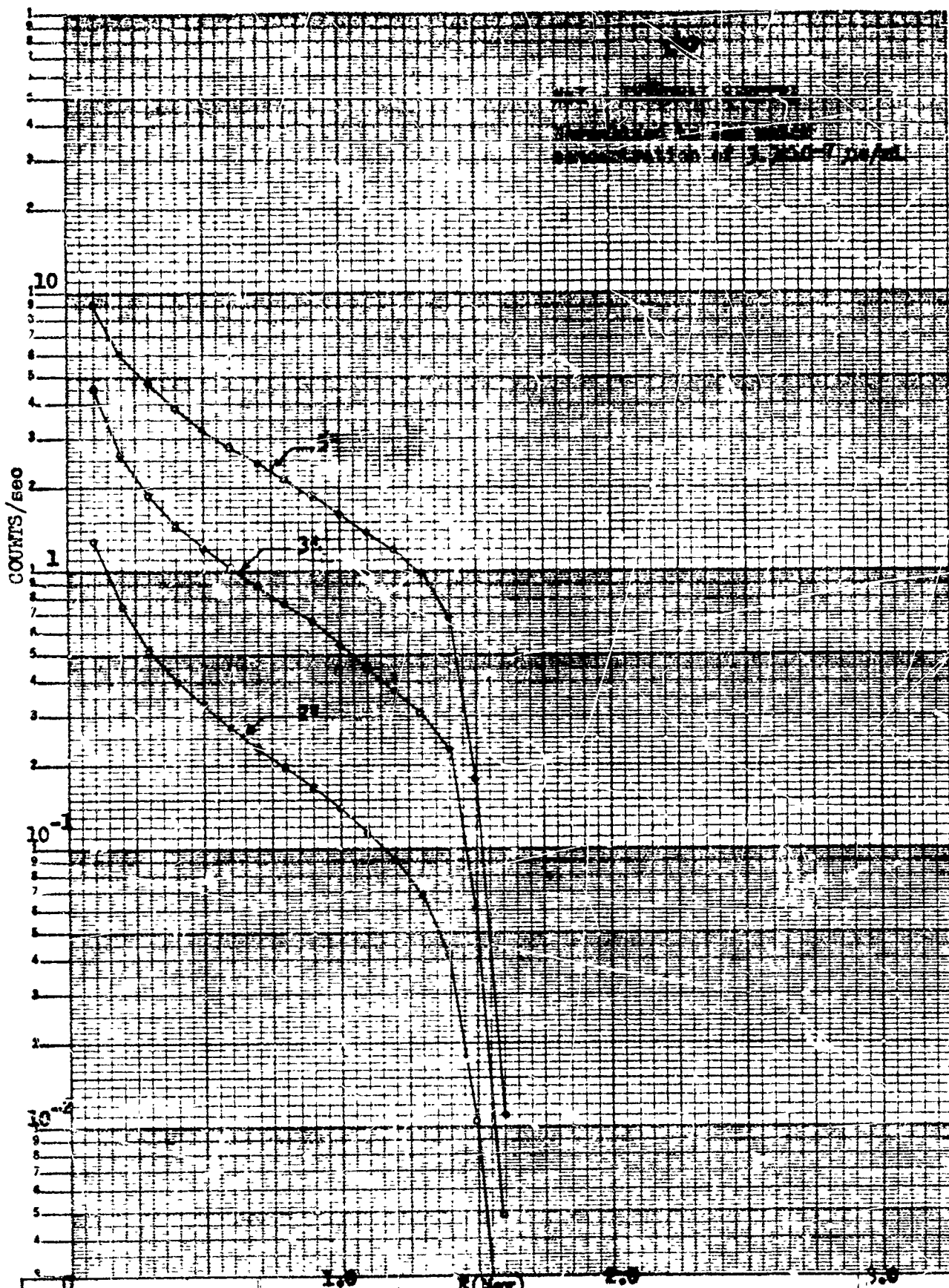
$\overline{N(E)}$ COUNTS/sec/Mev



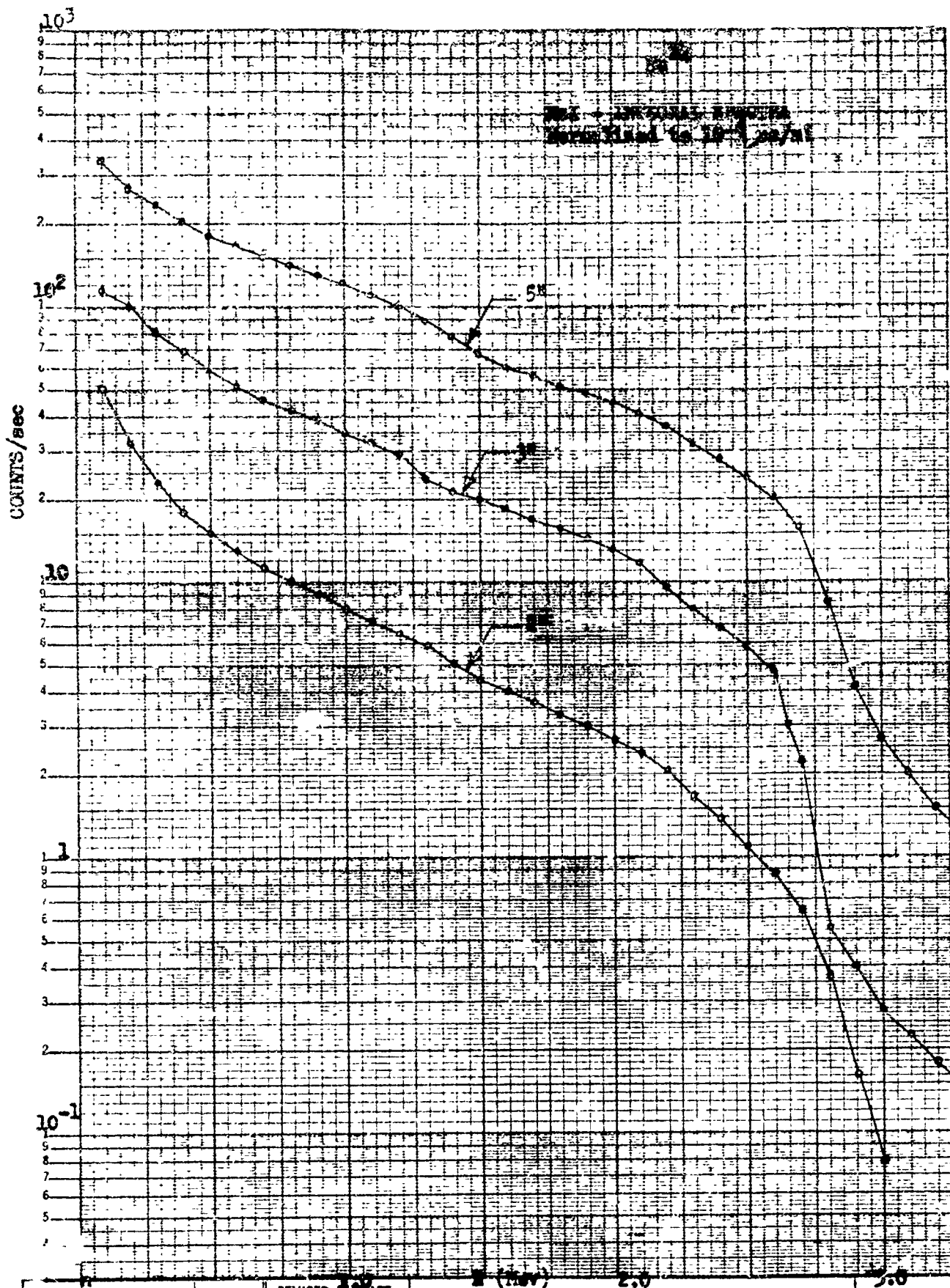
CALC		REVISED	DATE	E(Mev)	3.0
CHECK				Figure 38. Na ²⁴ Differential Spectra	D2-7908
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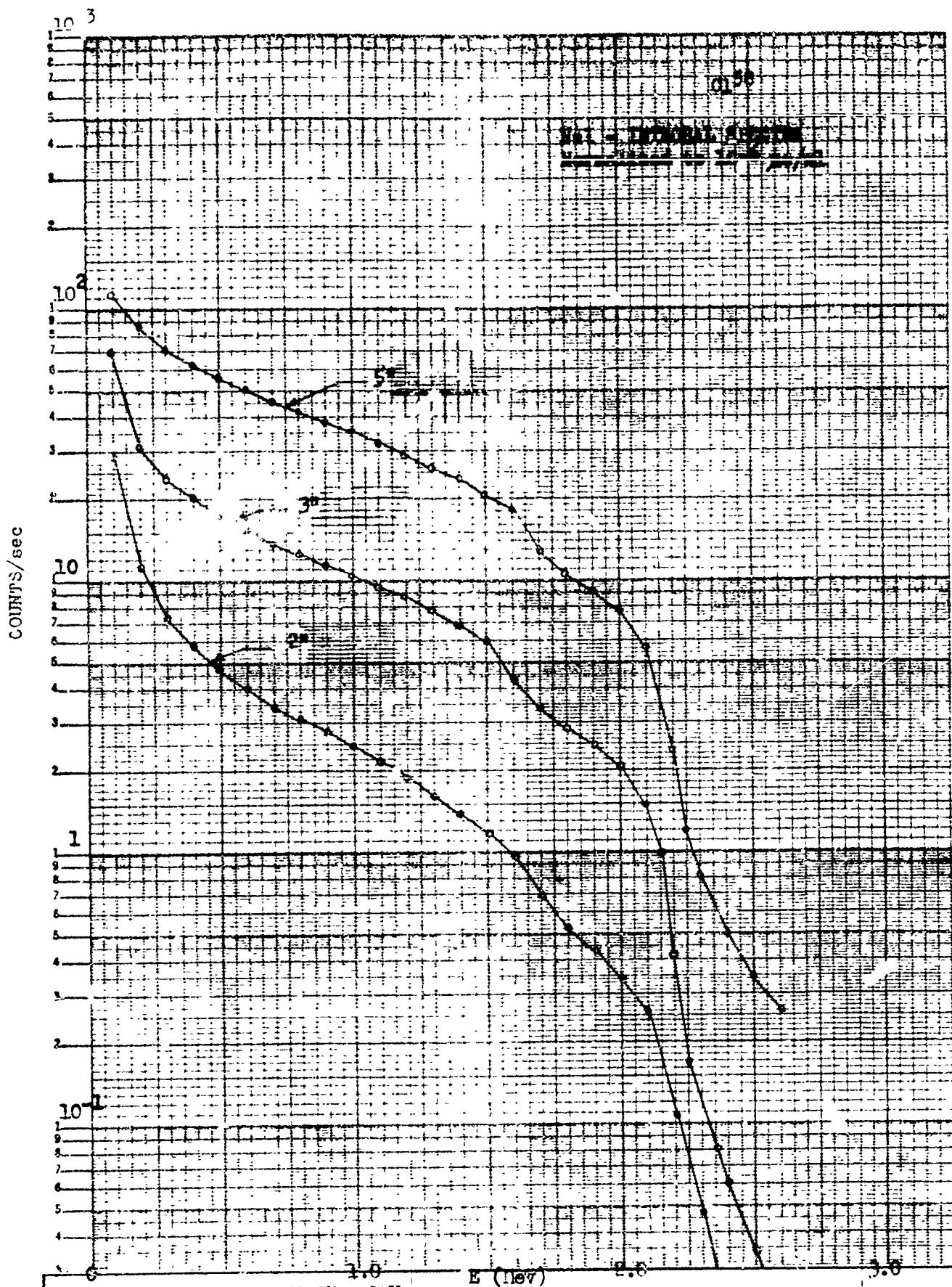
CALC		REVISED	DATE	Figure 39. Cl^{38} Differential Spectra for Pilot B Plastic Phosphors BOEING AIRPLANE COMPANY	D2-7908
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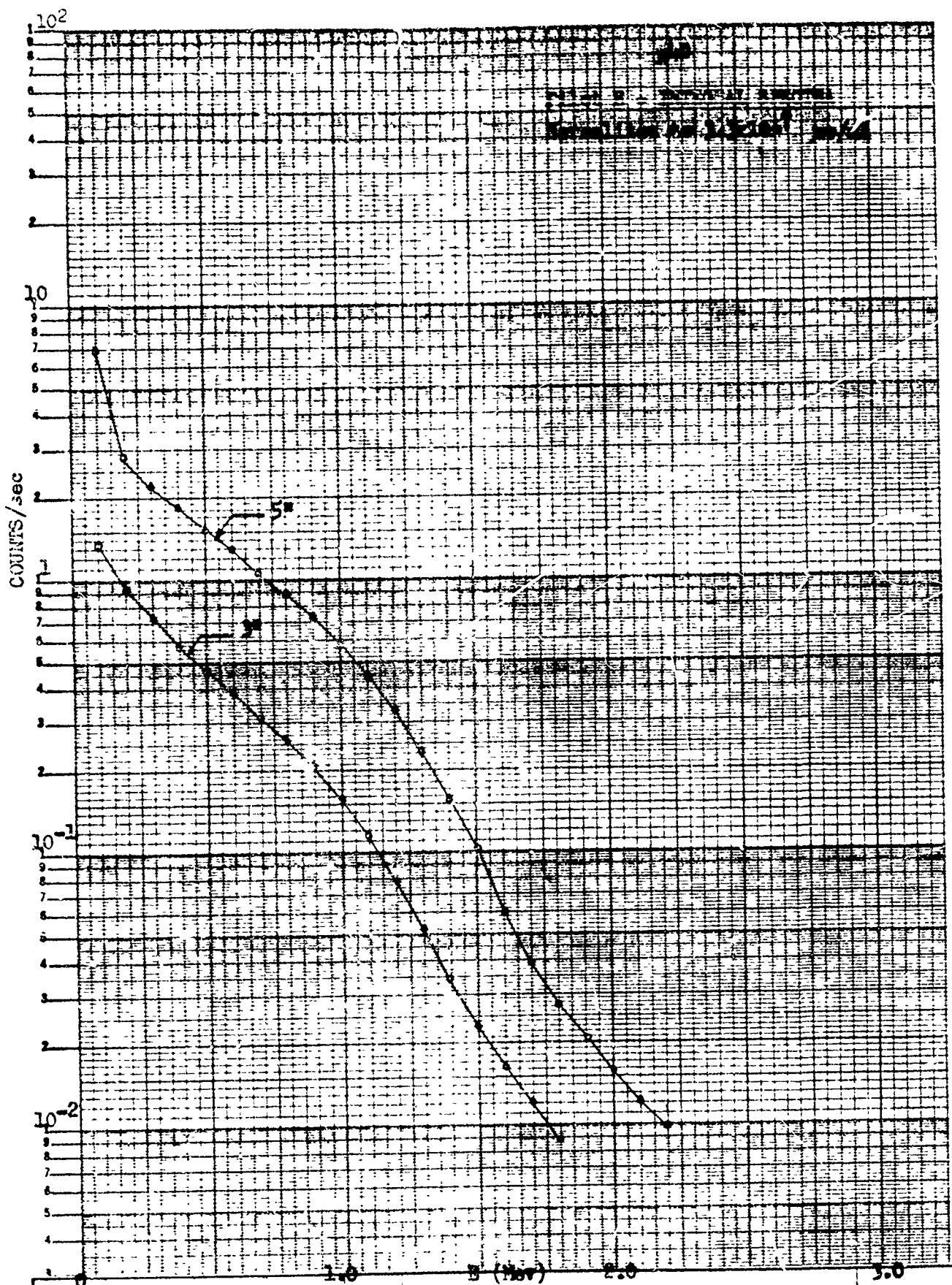
CALC CHECK APR APR	REVISED DATE	Figure 40. K^{40} Integral Spectra for NaI(Tl) Crystals BOEING AIRPLANE COMPANY	D2-7908 Sec. 2. PAGE 01 OF
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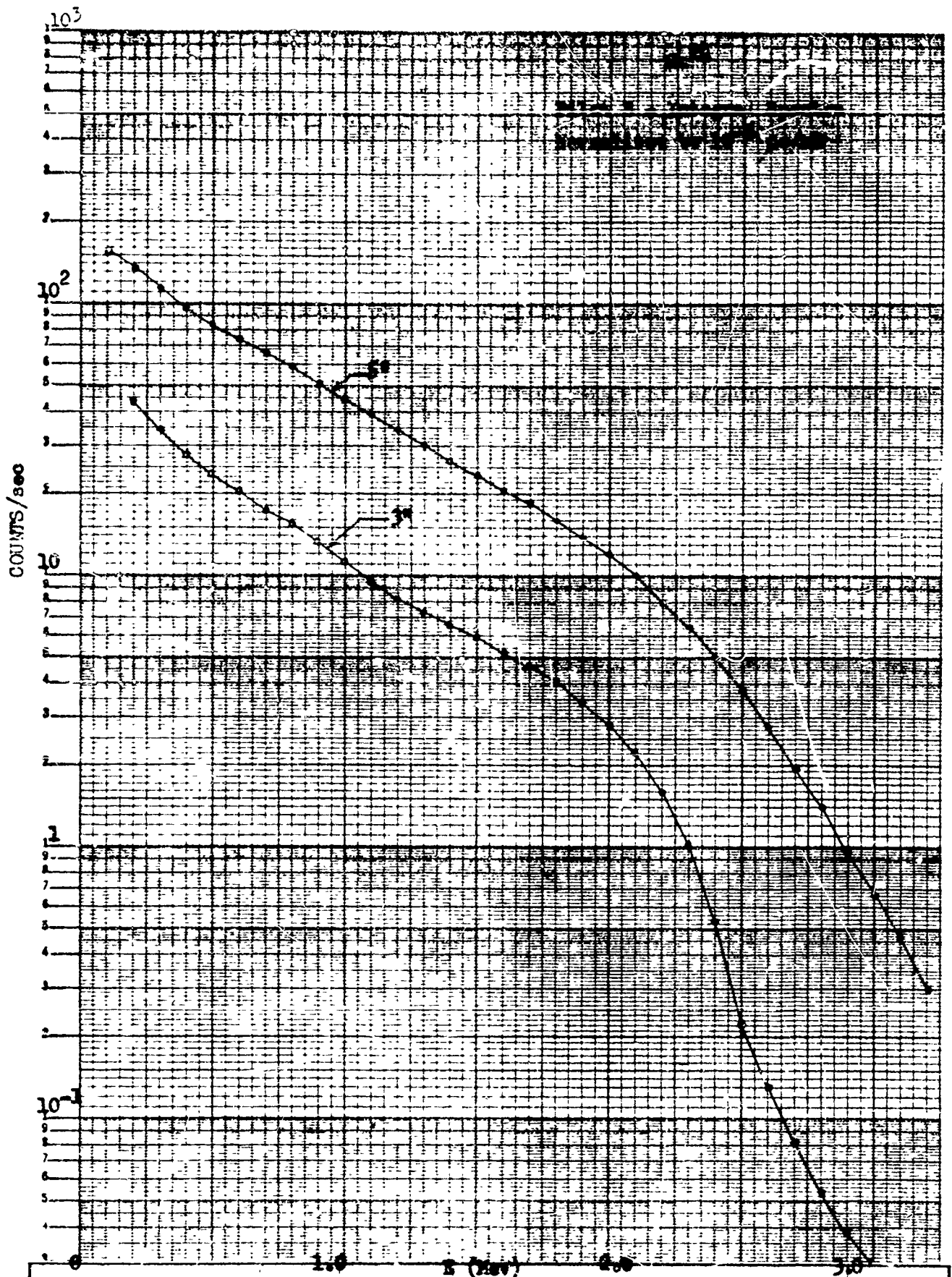
CALC		REVISED	DATE	Figure 41. Na^{24} Integral Spectra for NaI(Tl) Crystals BOEING AIRPLANE COMPANY SEATTLE 14, WASHINGTON	D2-7908
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CALC CHECK APR APR	REVISED DATE	Figure 42. Cl^{38} Integral Spectra for $\text{NaI}(\text{Tl})$ Crystals BOEING AIRPLANE COMPANY	D2-7908 Sec. 2. PAGE 93 OF
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CAIC	REVISED	DATE	Figure 43. K^{40} Integral Spectra for Pilot B Plastic Phosphors	D2-7908
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Figure 44. Na²⁴ Integral Spectra for
Pilot B Plastic Phosphors

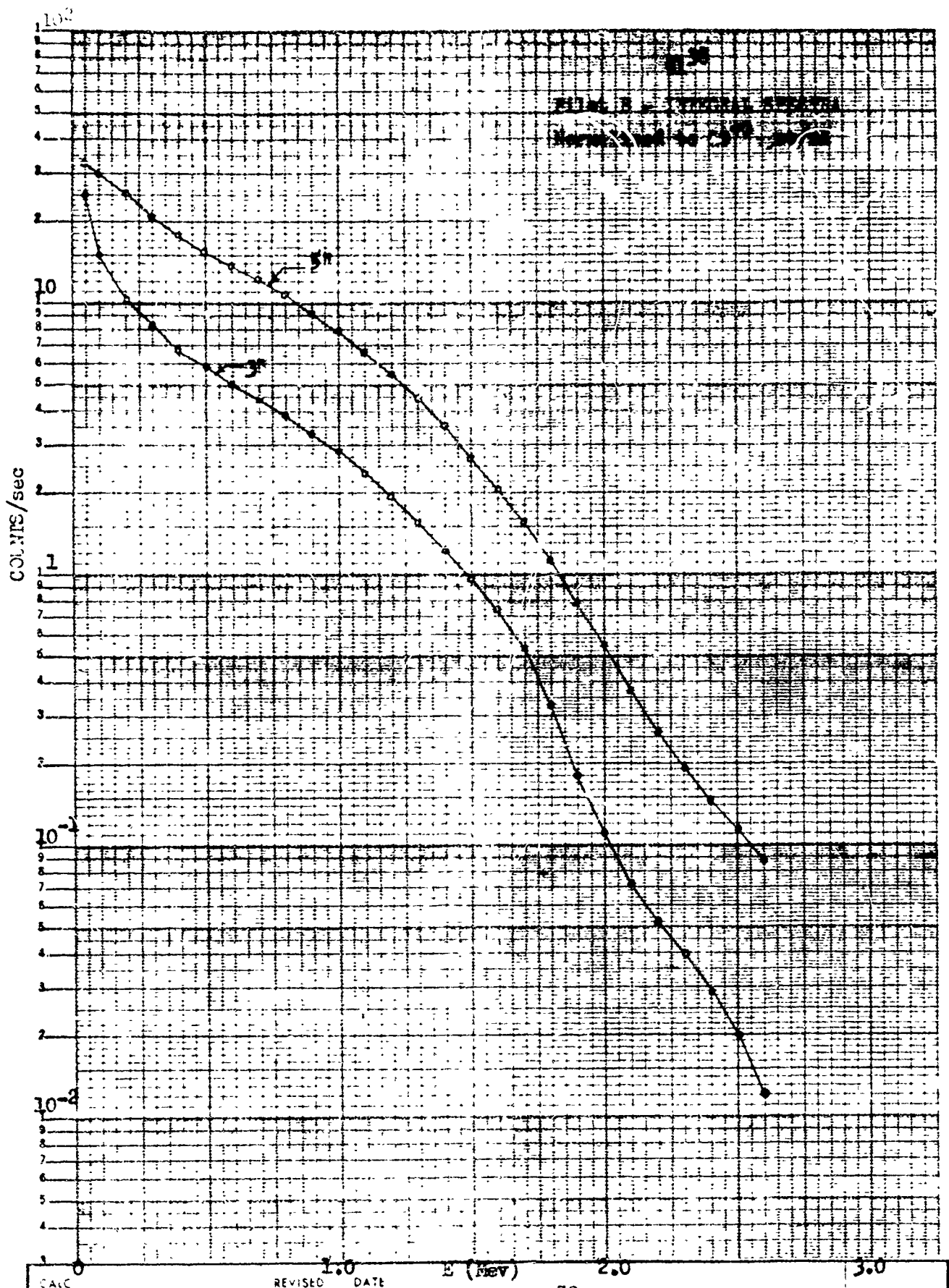
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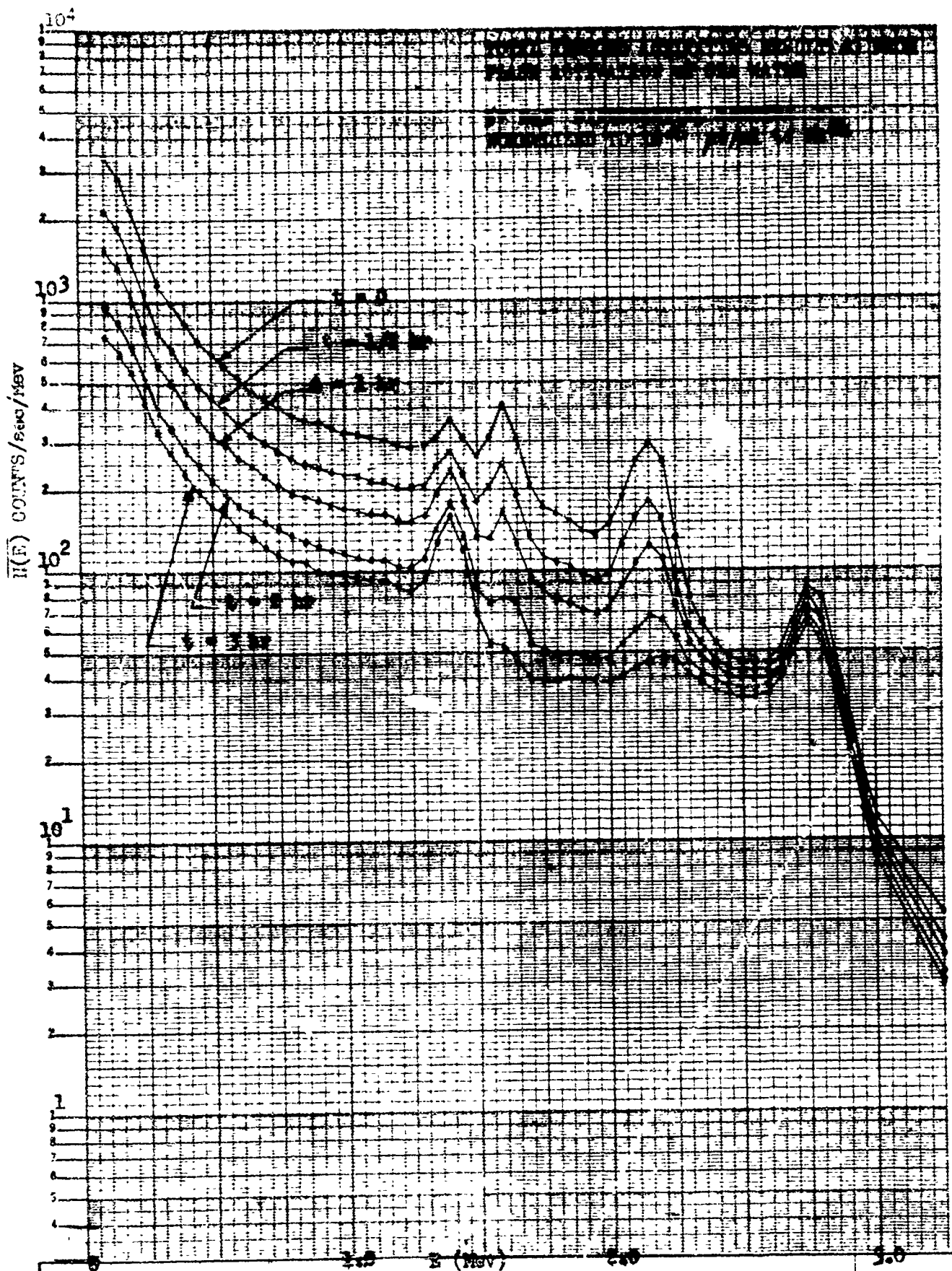
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CALC CHECK APR APR	REVISED DATE	Figure 45. Cl^{38} Integral Spectra for Pilot B Plastic Phosphors BOEING AIRPLANE COMPANY	D2-7908 Sec. 2. PAGE 96 OF
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CALC _____ CHECK _____ APR _____ APR _____	REVISED _____ DATE _____	Figure 46. Total Activity at Various Times After Flush Activation of Sea Water. BOEING AIRPLANE COMPANY	D2-7908 Sec. 2. PAGE 97 OF
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is the ratio obtained from flash activation of sea water. The change in the shape of the spectrum with time is then due to the difference in the decay rates of the chlorine-38 (half life 37 min.) and sodium-24 (half life 15 hr.). The component spectra together with the sum spectrum at $t = 0$ and $t = 3$ hr. are shown in Figures 47 and 48 which show the relative contributions due to the sodium-24 and chlorine-38 at these times. (C)

In Figures 49 to 53 the sodium-24 and chlorine-38 spectra are combined with the spectrum of potassium-40 for the activities predicted for various values of neutron leakage flux. These spectra give the expected spectral shape in sea water immediately after the passage of the submarine. These are spectra only of the activities present in sea water, other background activities are not included. The initial sodium-24 and chlorine-38 activities were obtained from the analysis given in the previous section and correspond to a hull area of 600 square feet and a submarine speed of 20 knots. The neutron leakage fluxes which were used together with the resulting activities are listed in Table 8. (C)

7.3 COUNTING EFFICIENCY FOR VOLUME DISTRIBUTED SOURCES

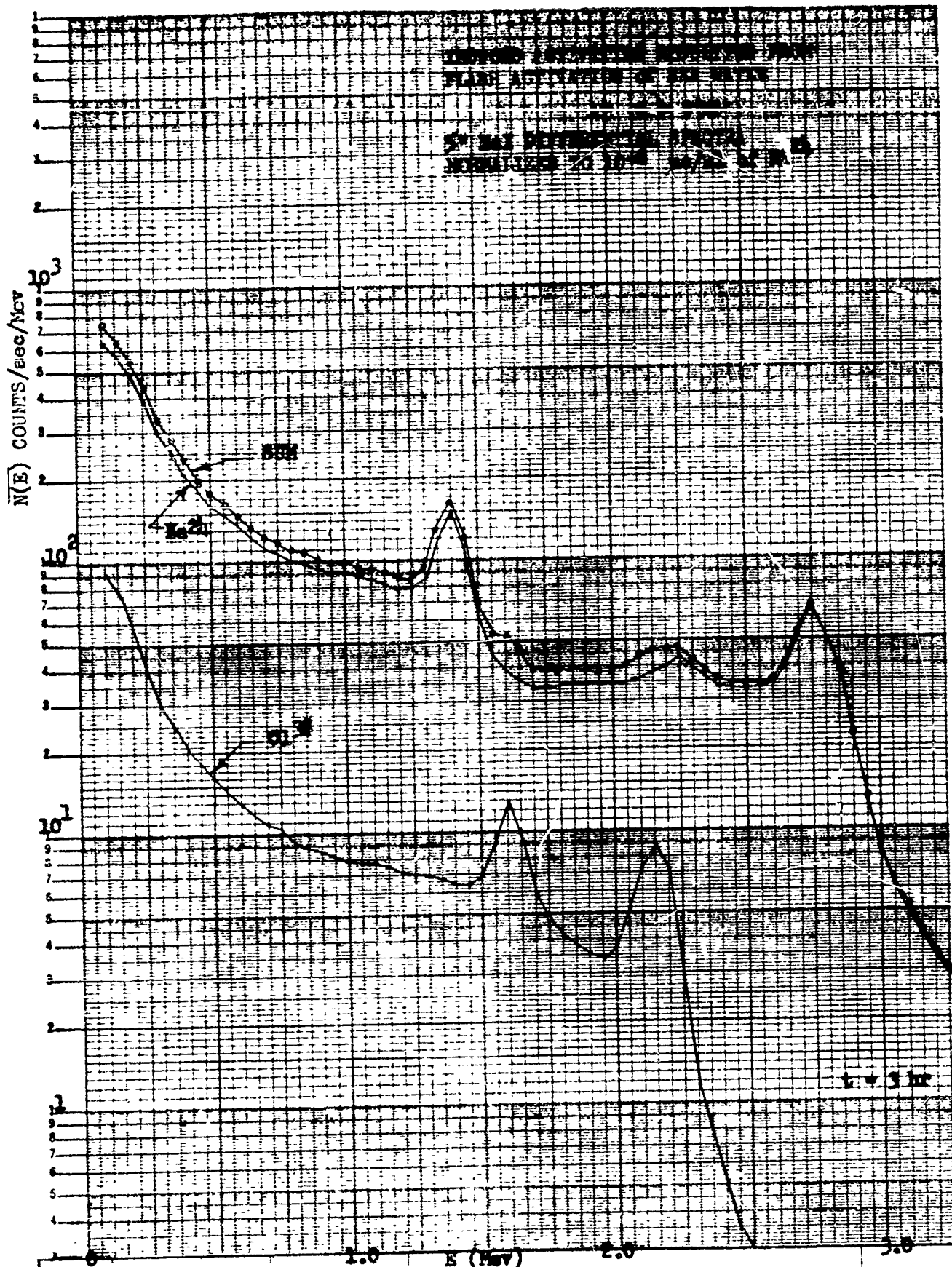
a. The counting rate in a detector from volume distributed sources arises from gamma rays of two origins. First, the primary gamma rays which are a direct result of the decay, and second, the buildup gamma rays which result from multiple scattering of the primary gamma rays by the medium. (U)

If one neglects the contribution from the buildup, the counting rate due to the primary gamma rays can be easily estimated. Let (U)

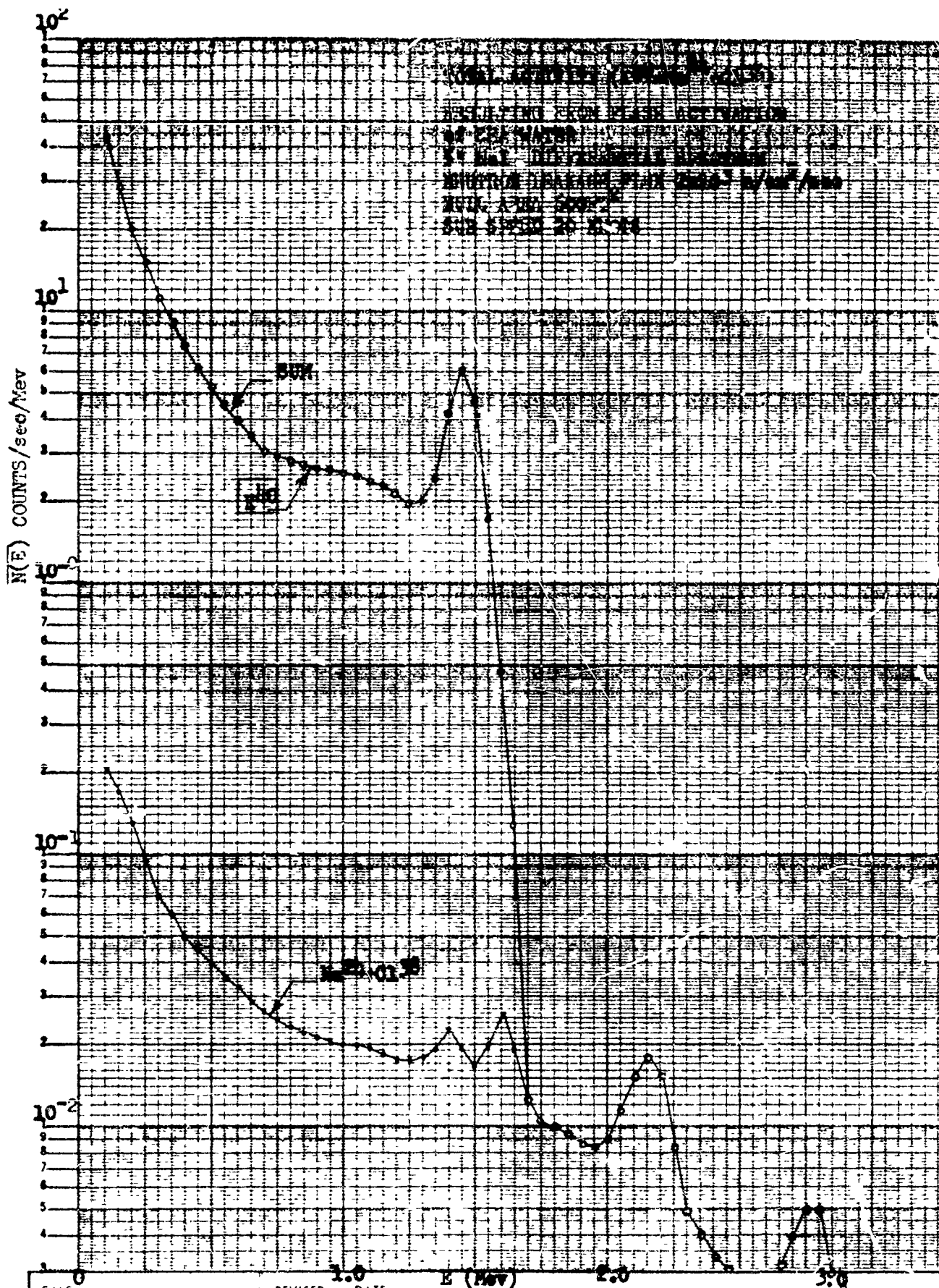
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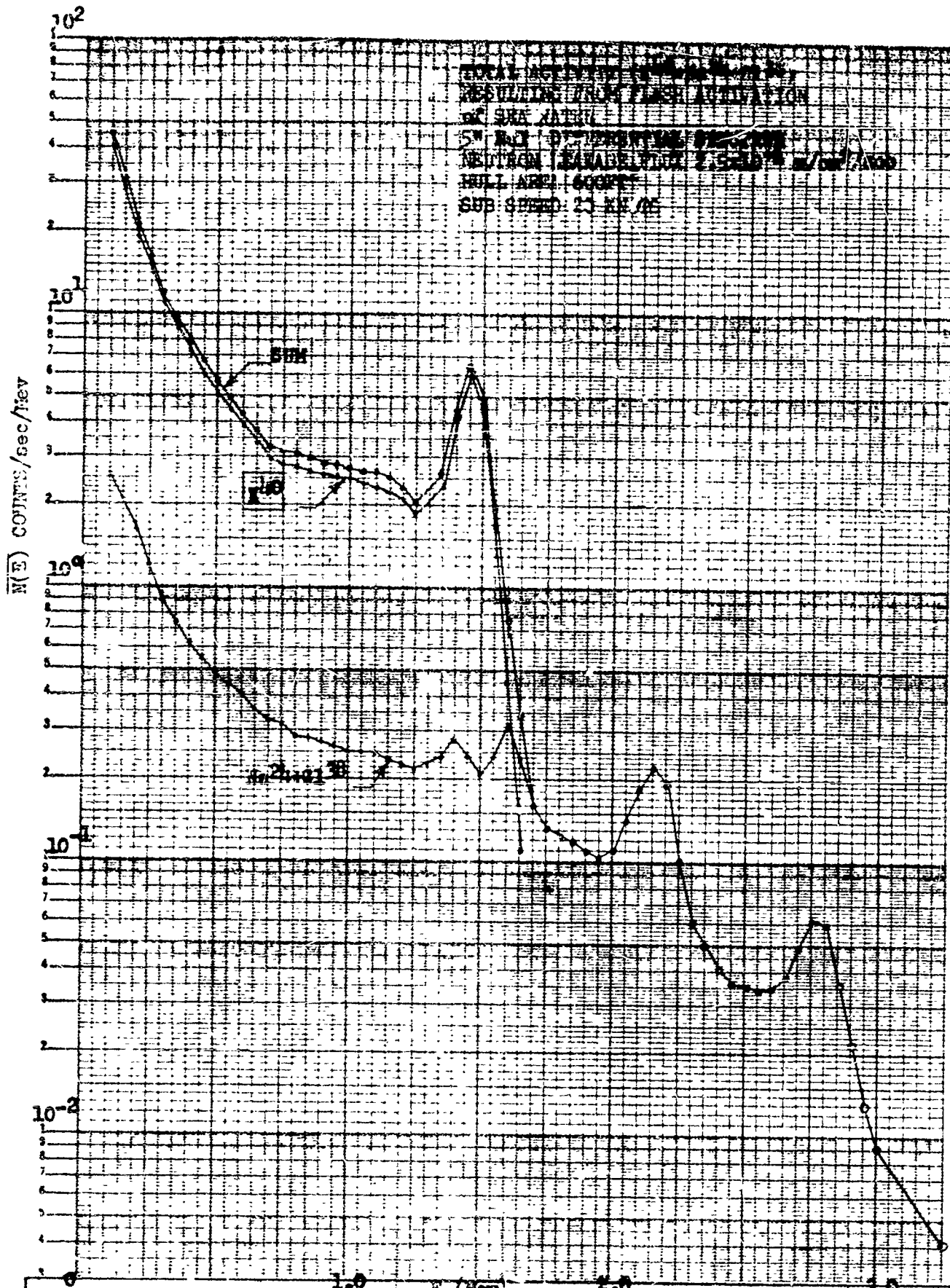
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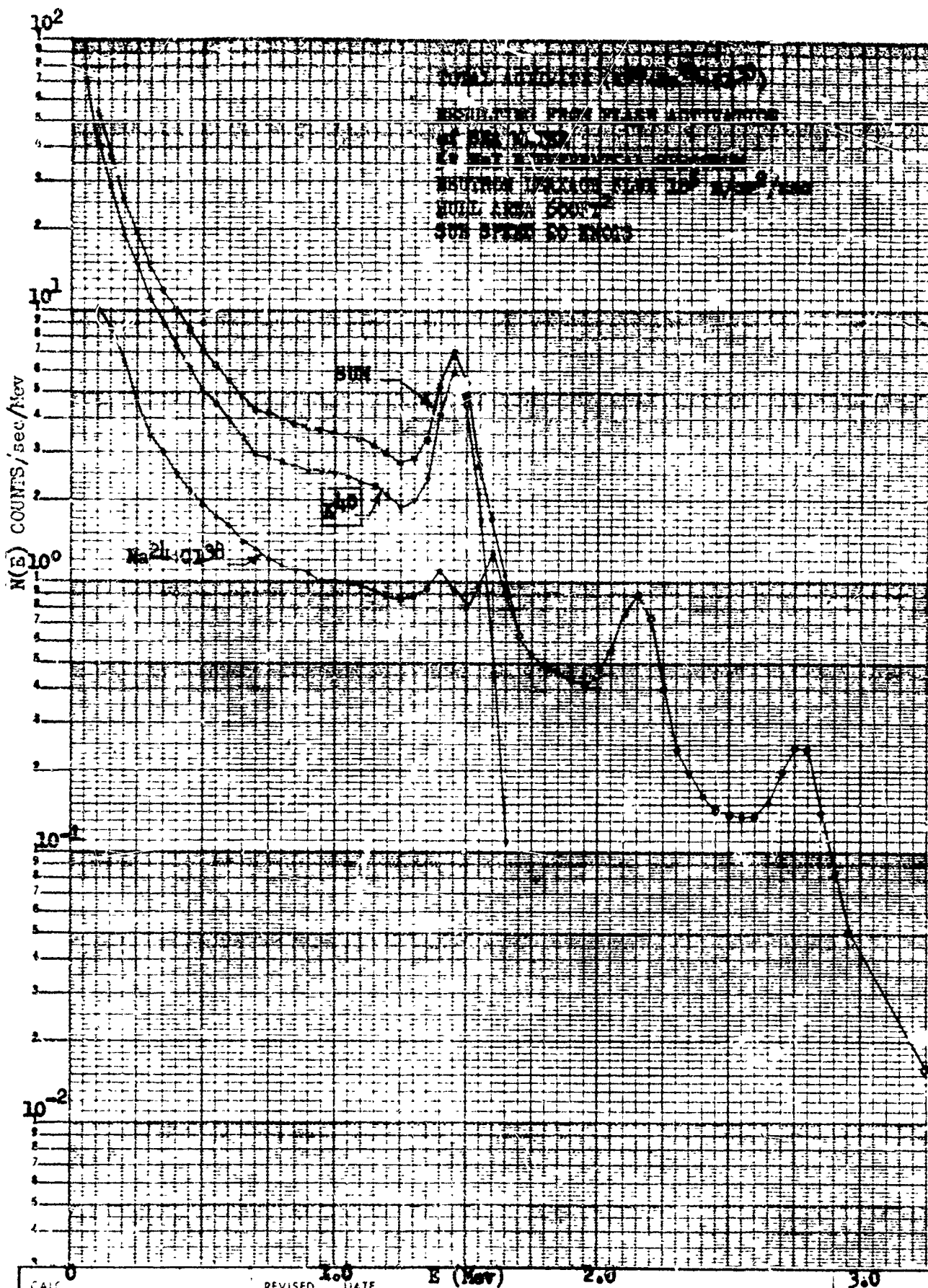
CALC CHECK APR APR	REVISED DATE	1.0 E (Mev) 2.0 3.0	Figure 48. Cl^{38} and Na^{24} Activities After Flash Activation of Sea Water, $t = 3$ hours BOEING AIRPLANE COMPANY	D2-7908 Sec. 2. PAGE 100 OF
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CALC CHECK APR APR	REVISED DATE	Figure 49. Activities Resulting from Flash Activation of Sea Water with Ther- mal Neutrons. Flux = 2×10^3 n/cm ² /sec. BOEING AIRPLANE COMPANY	D2-7908 Sec. 2. PAGE 101 OF
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CALC _____ CHECK _____ APR _____ APR _____	REVISED _____ DATE _____	E (Mev) _____ Figure 50. Activities Resulting from Flash Activation of Sea Water With Ther- mal Neutrons. Flux = 2.5×10^4 n/cm ² /sec. BOEING AIRPLANE COMPANY	D2-7908 Sec. 2. PAGE 102 OF
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CALC CHECK APR APR	REVISED DATE	Figure 51. Activities Resulting from Flash Activation of Sea Water With Ther- mal Neutrons. Flux = 10^5 n/cm ² /sec. BOEING AIRPLANE COMPANY	D2-7908 Sec. 2. PAGE 103 OF
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TABLE 8. Summary Neutron Flux and Induced Activity at $t = 0$.

<u>Neutrons/cm²/sec</u>	<u>Sodium-24 Specific Activity</u>	<u>(Na²⁴ + Cl³⁸) Specific Activity</u>
2×10^3	$6.08 \times 10^{-11} \mu\text{c/ml}$	$5.1 \times 10^{-10} \mu\text{c/ml}$
2.5×10^4	$7.6 \times 10^{-10} \mu\text{c/ml}$	$6.38 \times 10^{-9} \mu\text{c/ml}$
10^5	$3.04 \times 10^{-9} \mu\text{c/ml}$	$2.55 \times 10^{-8} \mu\text{c/ml}$
10^6	$3.04 \times 10^{-8} \mu\text{c/ml}$	$2.55 \times 10^{-7} \mu\text{c/ml}$
10^7	$3.04 \times 10^{-7} \mu\text{c/ml}$	$2.55 \times 10^{-6} \mu\text{c/ml}$

the detector be considered a sphere of radius r_0 with total detection efficiency for the primary gamma ray. Let the specific activity of the source be N_0 and the detector efficiency be n , the total absorption coefficient of the medium be μ . Then the fraction of the gamma rays which are radiated by the volume element $da \cdot dr$ in the solid angle subtended by the detector is approximately:

$$\frac{\pi r_0^2}{4\pi r^2} = \frac{A_0}{4(4\pi r^2)}$$

where A is the surface area of the detector. Thus the total number of gamma rays detected per second from an infinite volume is:

$$\begin{aligned} N_\infty &= n \int_{r_0}^{\infty} \int_{4\pi\Omega} N \frac{A}{4(4\pi r^2)} e^{-\mu(r-r_0)} da dr \\ &= \frac{N_0 A}{4} \int_{r_0}^{\infty} e^{-\mu(r-r_0)} dr = \frac{N_0 A}{4} \int_0^{\infty} e^{-\mu r'} dr' \\ &= \frac{N_0 A}{4\mu} \end{aligned}$$

If the volume of the active medium is finite and is a sphere of radius R then the counting rate N_R would be:

$$N_R = \frac{N_0 A}{4\mu} (1 - e^{-\mu(R-r_0)})$$

If we take R equal to 40 inches, which is the closest distance from the detector to the outside of our tank, we have that:

$$\begin{aligned} \frac{N_\infty}{N_R} &= 1.02 \quad \text{with } \mu \cong 0.042 \text{ cm}^{-1} \\ &= 40 \text{ inches} = 100 \text{ cm} \\ & \quad r_0 \leq 6 \text{ cm} \end{aligned}$$

for the 2.75 Mev gamma ray of sodium-24. Thus we see that our tank was equivalent to an infinite volume to better than two percent for the highest energy gamma ray used in our experiment. For the other gamma rays (≤ 2.15 Mev) the error was less than one percent.

- b. In order to compare the relative efficiencies of the various detectors we see that if μ' is the linear absorption coefficient of the detector, I_0 the incident gamma ray flux, and I the flux of gamma rays which pass through the detector, then the detector efficiency η is given by:

$$\eta = \frac{I_0 - I}{I_0} = (1 - e^{-\mu' d})$$

where d is the diameter of the detector which is assumed to be a sphere. Also the gamma rays are assumed to arrive normal to the detector surface. For values of d such that $e^{-\mu d} \ll 1$, $\eta \cong 1$. For values of d such that $\mu d \ll 1$, $e^{-\mu d} \cong 1 - \mu d$ and $\eta \cong \mu d$. Making use of the equation for the detector counting rate we see that for d large ($e^{-\mu d} \ll 1$), N_0 is proportional to the area; while, for d small ($\mu d \ll 1$), N_0 is proportional to the volume. This is reasonable for it states that if the detector is large compared with the mean free path of the gamma ray (i.e. $e^{-\mu d} \ll 1$), then all of the incident gamma rays are counted and their number depends on the surface area. On the other hand if the crystal is small compared with the mean free path ($\mu d \ll 1$) then the number absorbed depends only on the volume of absorbing material.

The linear absorption coefficient as a function of energy is plotted in Figure 54 and NaI(Tl) and in Figure 55 for the Pilot B plastic phosphor. To delineate the regions for which the crystal area and crystal volume are important for phosphors of various sizes and types, values of constant μd as functions of energy and phosphor diameter are shown in Figures 56 and 57. The values

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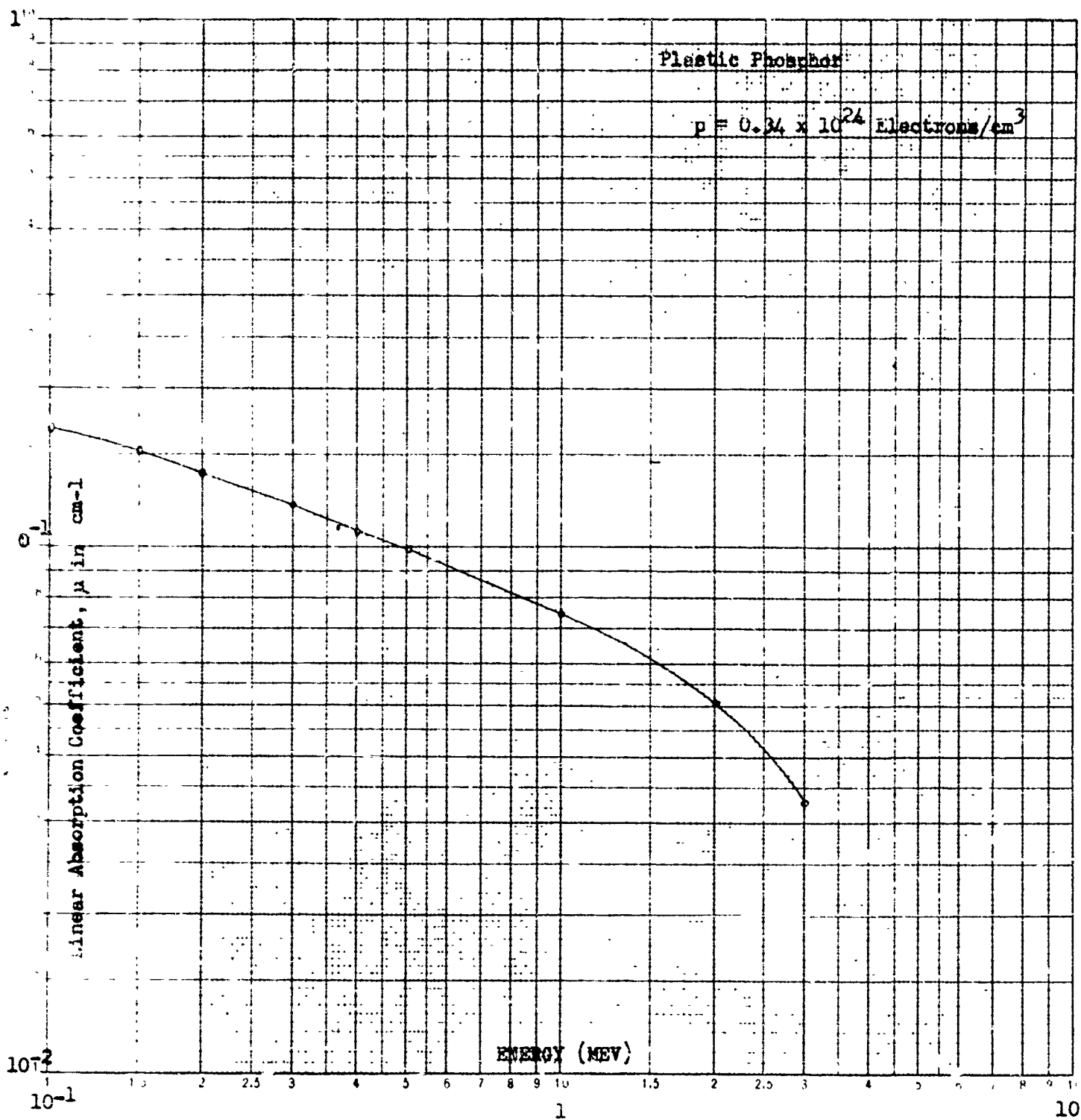
TOTAL LINEAR ABSORPTION COEFFICIENT of NaI (Tl)

Total Linear Absorption Coefficient, μ in cm^{-1}

10⁻¹
0.1

ENERGY (MEV)

CALC.			REVISED	DATE	Figure 54. Total Linear Absorption Coefficient for Sodium Iodide (NaI (Tl) crystal). BOEING AIRPLANE COMPANY SEATTLE 24, WASHINGTON	D2-7908
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CALC.			REVISED	DATE	Figure 55. Total Linear Absorption Coefficient for Plastic Phosphors such as the Pilot "L".	D2-7906
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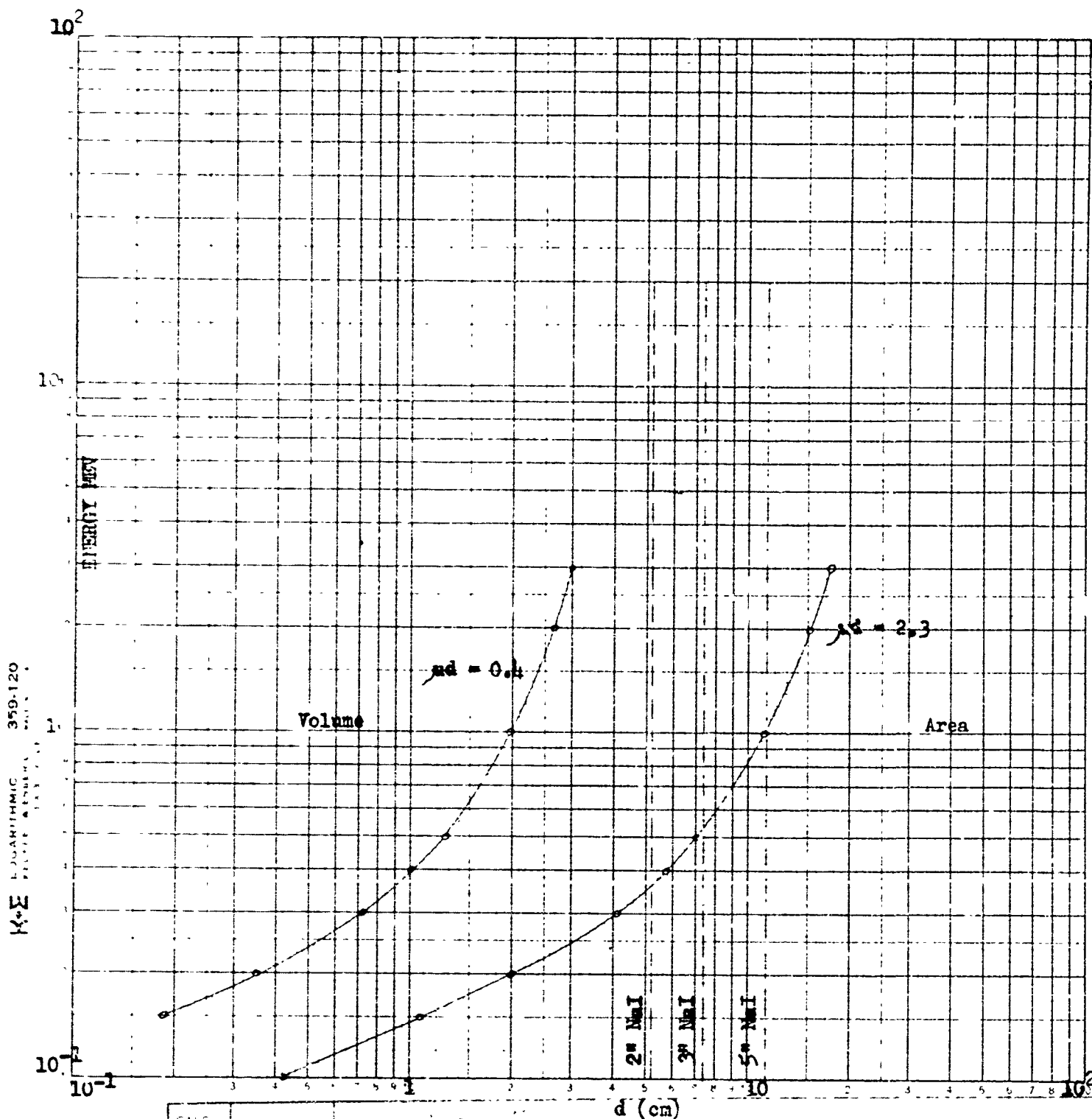


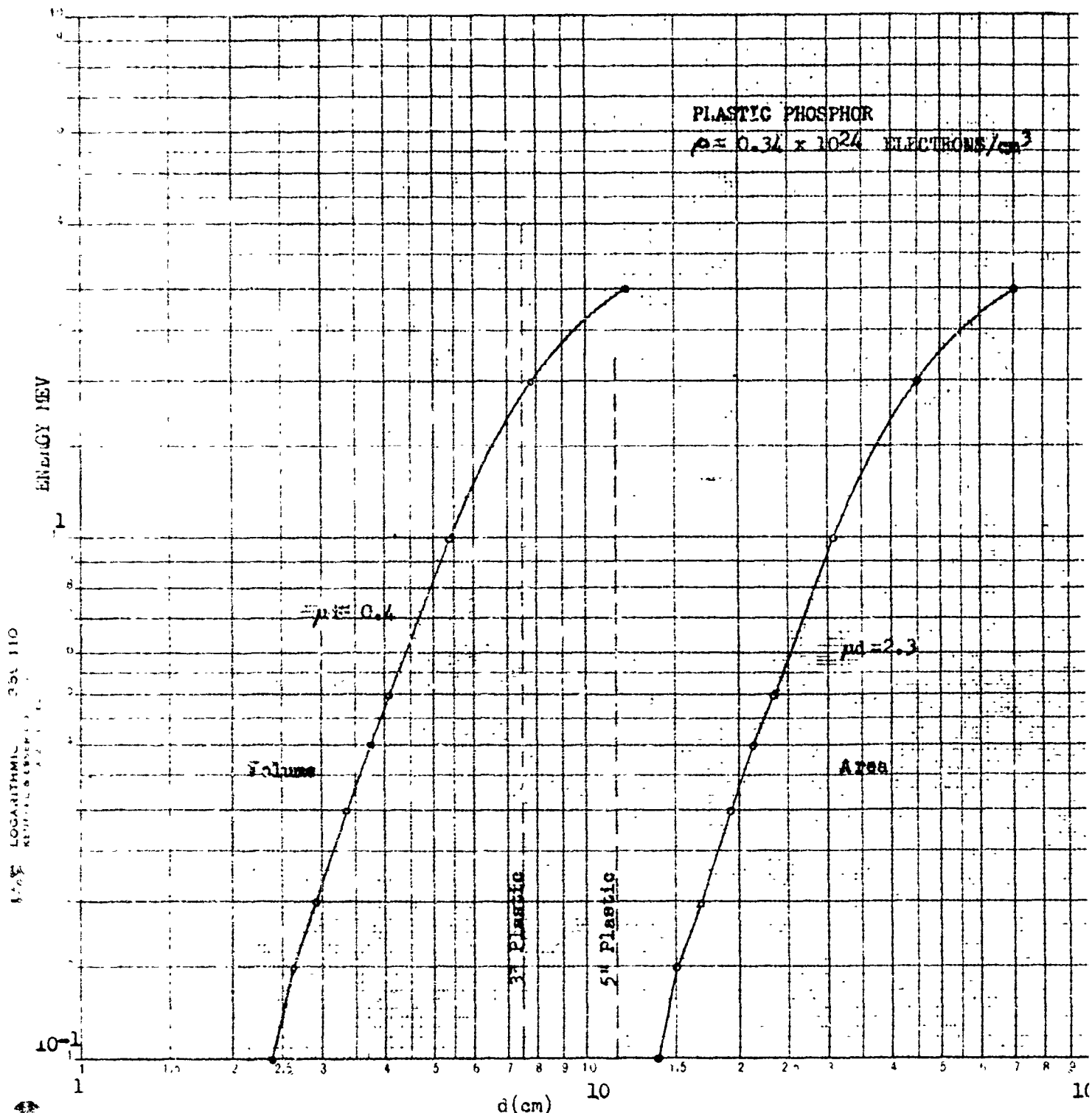
Figure 56. Plots Showing Transition from Volume to Area Response for Tellurium Activated Sodium Iodide Crystals.

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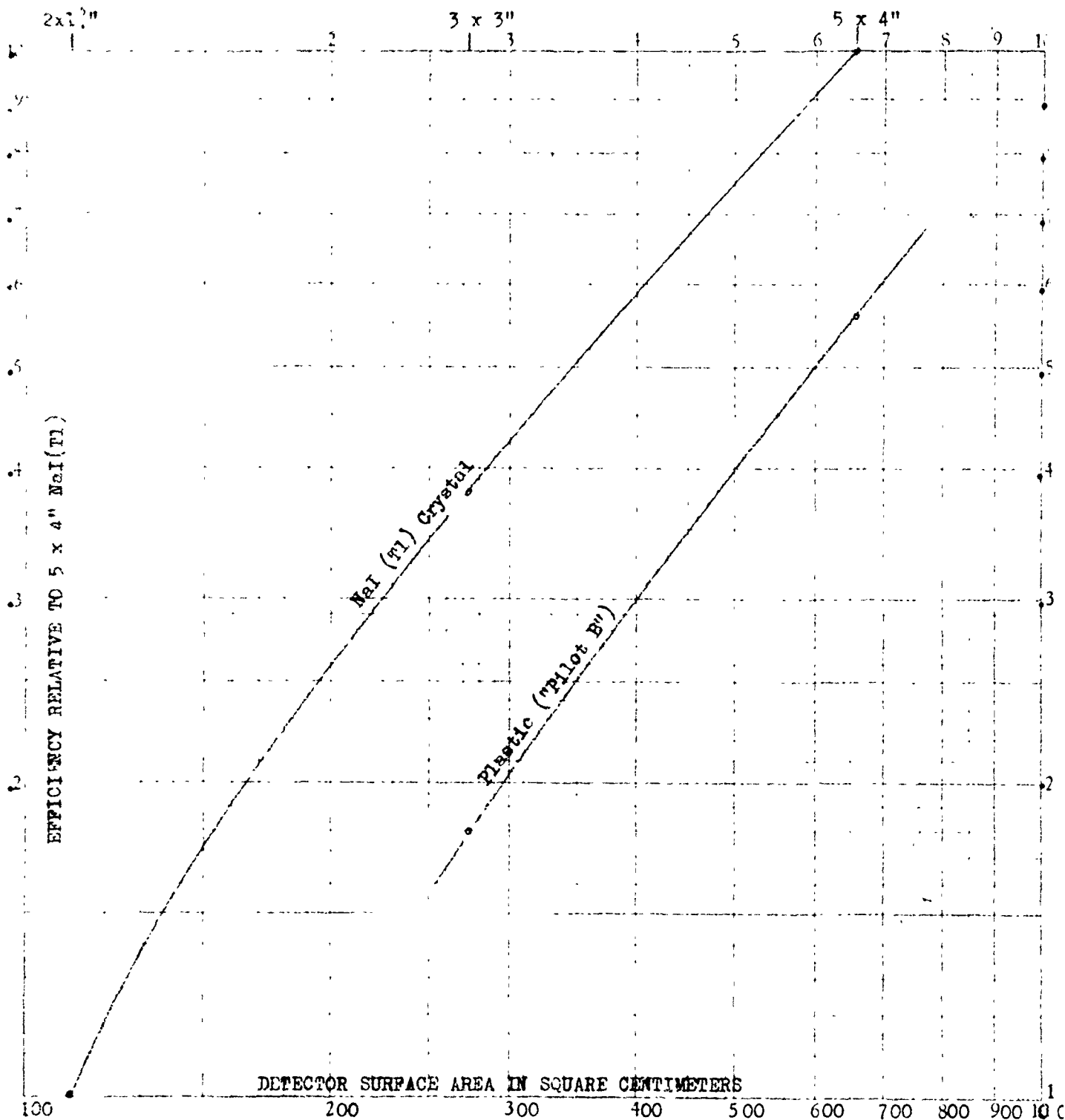


CALC.		REVISED	DATE	Figure 57. Plots Showing Transition from Volume to Area Response for Plastic Phosphors such as the Pitot "B".	D 2-7908
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of μd chosen were $\mu d = 0.4$ which gives the counting rate as proportional to the volume within 10 percent and $\mu d = 2.3$ for which the counting rate is within 10 percent proportional to the area. Also indicated on the curves are the values of d corresponding to the mean diameters of the crystals used in the tests. As can be seen, for not too large energies, all of the NaI crystals fall within or near the region of area dependence, while the plastic phosphors fall in the region where no prediction can be made.

The relative efficiencies of NaI(Tl) and plastic phosphors for integral counting above 250 kev are plotted in Figure 58 as a function of the detector area. All of the efficiencies are referred to the 5 x 4 inch crystal. Comparative tests are being made with larger plastic phosphors and with a liquid scintillant in various size containers. Discussion of Figure 58 will be deferred until the new data become available.

DETECTOR SIZE IN INCHES



CALC			REVISED	DATE	Figure 58. Relative Efficiency vs. Surface area for Integral Counts Above 250 kev Compared to 5x4" NaI(Tl) BOEING AIRPLANE COMPANY	D2-7908
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INDUCED RADIOACTIVITY IN SEA WATER

PREPARED BY Leo diRicco

SUPERVISED BY W. E. Nesbitt

APPROVED BY D. A. Hicks (DATE) _____

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1. OBJECTIVE

The objective of this section is to present a summary of the chemical means by which radioactivity artificially induced into sea water may be concentrated for detection. An analysis of the principle constituents of sea water will be made in order to establish the most practical medium on which to detect. Further, an evaluation is to be made of methods believed to warrant exploratory laboratory investigation from which basic data for firm engineering and cost estimates may be made.

2. VAPORIZATION TECHNIQUES

2.1 INTRODUCTION

The process of transferring water molecules from the liquid phase to the vapor phase is called vaporization. A direct method for the concentration and recovery of the solids dissolved in sea water is by removal of the water through vaporization--since the dissolved solids are non-volatile at the vaporization temperature of water, they remain behind and can be recovered. The recovery of dissolved solids from sea water by vaporization techniques is basically a problem of supplying the latent heat of vaporization to the fluid and of providing means for recovery of the solids. This is an old concept and is reflected in the variety of equipment which is in use for this purpose today. Information and requirements data for vaporization processes for the concentration of solids dissolved in sea water are presented in the following pages.

2.2 EVAPORATORS

Evaporators are intended to concentrate the induced radioactivity in sea water by evaporation to a concentrated solution.

2.2.1 BACKGROUND

Since the task to be performed is that of concentrating a solution by evaporating part or all of the water, the basic thermodynamic requirements for energy input will be the same for a given set of boundary conditions (the dissolved solids specific activity, dissolved solids content, and sample size), so that the preliminary development of this technique for our purposes is of necessity based on the operational characteristics and costs of the different kinds of equipment available for performing this operation. This assumes that the quantity of energy required for the evaporation of one pound of water is approximately the same for all conventional evaporators having the same number of stages. This is a valid assumption, accurate to within a few percent for a given set of boundary conditions.

2.2.2 TYPES (Ref. 1)

2.2.2.1 SINGLE EFFECT

A single effect evaporator is essentially a fluid container with a means for adding heat energy to the fluid and for removing the vapor. These are arbitrarily classified as:

- a. Apparatus using solar heat
- b. Apparatus heated by direct fire
- c. Apparatus with heating medium in the jacket
- d. Steam heated evaporators with tubular heating surfaces
- e. Miscellaneous

The mechanical construction of single effect evaporators is quite varied. Typical mechanical features and costs are illustrated in the literature (Refs. 2, 3, 4, 5).

2.2.2.2 MULTIPLE EFFECT

Multiple effect evaporators consist of a series of evaporator bodies so constructed that the vapor from one body is the heating medium for the next. The purpose of a multiple effect evaporator is to reduce the energy consumption for a given amount of evaporation.

If it is assumed that the terminal temperatures are the same for both single and multiple effect evaporator operations, then passing from a single effect evaporator to a multiple effect evaporator does not increase the capacity of an evaporator but it does (theoretically) increase the actual pounds of water evaporated per unit of energy input by a factor equal to the number of stages. Hence, passing from single effect to multiple effect evaporation techniques decreases energy cost but increases apparatus cost. The optimum number of stages to be used must be determined for each application.

2.2.2.3 MISCELLANEOUS

Several novel schemes and equipment configurations are itemized below (Ref. 5):

- a. Centrifugal vapor compression
- b. Supercritical pressure distillation
- c. Flash distillation technique

The equipment called for in these schemes falls into the category of single effect evaporators. The items are in a preliminary state of development and were not considered suitable for further evaluation at this time.

2.2.2.4 CONCLUSIONS

The use of a multiple effect evaporator appears to be unsuitable due to the lengthy "in process" time and "mixing" which occurs in the different effects. These factors would present problems of dilution and response during a "search." Further evaluation will therefore be limited to consideration of evaporative methods which are capable of a short response time of one minute or less. Such methods should assure that the amount of material "in process" will always be at a minimum.

2.2.3 PERFORMANCE CRITERIA

The following scheme proposes that a sea water sample be concentrated by evaporation to the point of solids saturation solubility. Such a method would have the advantage of complete liquid phase operation. The saturation solubilities for the major constituents dissolved in sea water at 20 degrees centigrade are as follows (Ref. 6):

<u>Component</u>	<u>(Solubility in Water (Grams/100 grams water))</u>	
NaCl	35.7	(85 percent of total)
KCl	34.7	(10 percent of total)
MgCl	35.3	(Slight)
CaCl ₂	59.5	(Slight)

Since the sodium chloride constitutes the preponderance of the dissolved solids, for the purpose of this evaluation the saturation solubility for all solids dissolved in sea water at 20° C was taken as 35.6 grams of dissolved solids per 100 grams of water. On this basis therefore, the quantity of water required to be evaporated in order to produce the required amount of "concentrate" for 200 counts per minute at a given sodium-24 and chlorine-38 activity and at a given detector efficiency can be obtained directly from Figure 1.

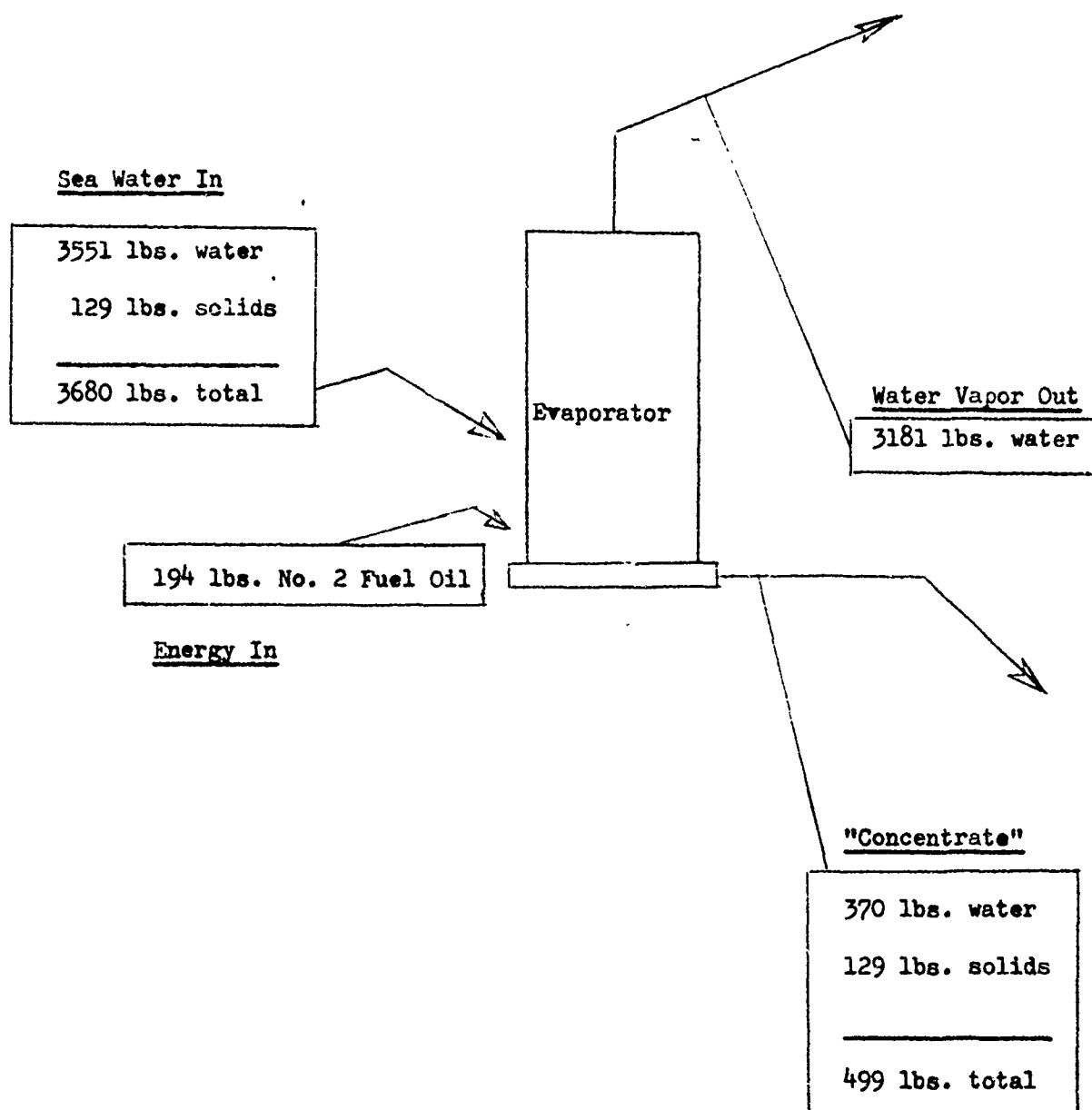


FIGURE 1. Example Establishing the Relationships of Input and Output for an Evaporator. Based on an activity of 10^{-9} microcuries per milliliter and 200 counts per minute (Figure 2).

2.2.3.1 SYSTEM CONSIDERATIONS

Computed system requirements for 200 counts in 1, 5, and 10 minutes are tabulated in Table 1 and plotted in Figure 2 (50 percent confidence level). Concentrating to a saturated solution gives a weight basis concentration ratio of 7.4 and a volume basis concentration ratio of 9.75.

This evaluation was based on computed detector efficiencies for the characteristic gamma emitted from sodium-24 and chlorine-38. The detector efficiency was based on the linear absorption coefficient in water at the full gamma energy.

2.2.3.2 EQUIPMENT CONSIDERATIONS

The following recommendation for equipment is based on two premises:

- a. Response time (material in process) to be held to a minimum--this in order to eliminate the "mixing" or "dilution" of samples during operation.
- b. Equipment to be of standard type insofar as standard indicates that a considerable history of experience exists for the item under consideration--this in order to eliminate at this time all intuitive and unproven schemes.

2.2.3.3 EQUIPMENT RECOMMENDATIONS

On the basis of the preceding discussion, the type of equipment recommended for the evaporation and concentration of sea water to a saturated solution is a Long-Tube Evaporator using forced circulation (Refs. 7, 8). The configuration may be either vertical or horizontal. The use of a circulating pump in this configuration minimizes scale formation by maintaining a high liquid velocity over the heat transfer surface.

TABLE 1. Detection of Sodium-24 and Chlorine-38 in Sea Water by Evaporation to a Concentrated Solution and Detecting with a 9-3/4 x 11 Inch Sodium Iodide Crystal.

Counts per Minute	Activity, in Microcuries per Milliliter	Sample Size in Liters and Pounds	Total Concentrate Volume Liters and Pounds	Water Evaporated (Pounds)	BTU per Sample at 1100 BTU per lb. Evaporated	Pounds of No. 2 Fuel Oil at 18,500 BTU per Pound
20	10 ⁻¹⁰	1920/4310	192/500	3810	4.14 x 10 ⁶	224
	10 ⁻⁹	30.6/69	3.1/10	59.1	6.4 x 10 ⁴	3.5
	10 ⁻⁸	< 10/ < 22	< 1/ < 3.2	< 19	< 21,000	< 2
	10 ⁻⁷	< 10/ < 22	< 1/ < 3.2	< 19	< 21,000	< 2
40	10 ⁻¹⁰	100,000/224,000	10,000/32,000	192,000	2.12 x 10 ⁸	1.15 x 10 ⁴
	10 ⁻⁹	56.5/127	5.6/17	110	1.21 x 10 ⁵	6.5
	10 ⁻⁸	< 10/ < 22	< 1/ < 3.2	< 19	21,000	< 2
	10 ⁻⁷	< 10/ < 22	< 1/ < 3.2	< 19	21,000	< 2
200	10 ⁻¹⁰	-----Not Possible-----				
	10 ⁻⁹	1640/3600	164/500	3100	3.6 x 10 ⁶	194
	10 ⁻⁸	30.6/69	3/10	59	6.4 x 10 ⁴	3.5
	10 ⁻⁷	< 10/ < 22	< 1/ < 3.2	< 19	< 21,000	< 2

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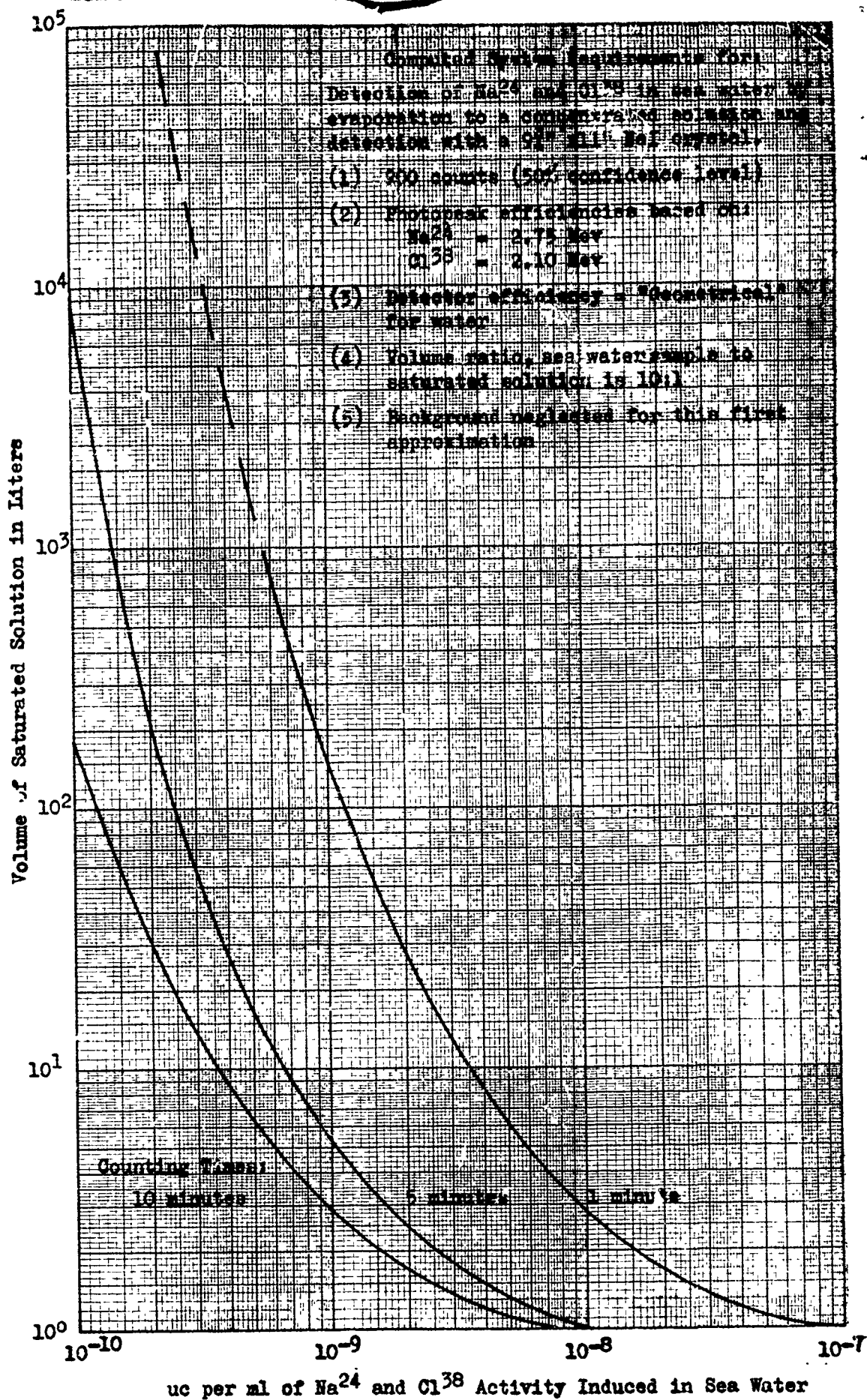


Figure 2. Computed System Requirements for 200 Counts in 1, 5, and 10 minutes.

2.2.3.4 SCALE FORMATION

In concentrating solutions such as sodium chloride, which have a solubility curve showing a very slight increase in solubility with temperature, the phenomenon of salting occurs (Ref. 1). A growth of crystals on the heating surface occurs which results in decreasing the heat transfer coefficient and consequently decreases the evaporator capacity. It is usually found that the beginning of salting occurs at a time when there has been some marked irregularity in the operation of the evaporator such as a decrease in steam pressure, increase in the boiling point, stoppage of circulation or any other factor that may cause a temporary decrease in boiling rate.

Scale formation may be minimized by maintaining a high liquid velocity across the heat transfer surface. Such action will prevent superheating the liquid and consequently will minimize scale formation.

2.2.4 CONCLUSIONS AND RECOMMENDATIONS

The detection of activity in sea water by evaporation of the sea water to a concentrated solution appears feasible for counting times of five minutes or longer (200 counts) at activity levels of 10^{-9} microcuries per milliliter or higher. For a counting time of 10 minutes or greater (200 counts) the detection of activity levels of the order of 10^{-10} microcuries per milliliter appears possible.

It is recommended that experimental work be carried out to establish the efficiency with which gamma radiation emanating from a saturated saline solution containing sodium-24 and chlorine-38 can be detected using a sodium iodide crystal in order to establish the mass absorption

coefficients for saturated saline solutions. This information is required in order to establish the necessary design parameters for a working system.

2.3 SPRAY DRIERS

Spray driers are intended to concentrate the induced radioactivity in sea water by evaporation to dry solids.

2.3.1 BACKGROUND (Ref. 9)

Spray drying is an evaporative method of concentrating solids in which a solution capable of being pumped or a suspension is sprayed into a stream of the drying medium--usually heated air. The large specific surface area of the small droplets allows the solvent to evaporate quickly, leaving a dry product in the form of a powder.

One of the basic characteristics of spray-drying is the extremely short "in process" (or retention) time for the material in question; the usual time interval during which the material is "in process" being 30 seconds or less. Because of this extremely short process time and fundamentally continuous mode of operation, the spray-drying technique has been chosen as the one offering the most potential for the evaporative-type concentration of the solids contained in sea water.

The essential components of a spray-drier are:

- a. Means of fluid dispersion.
- b. Contact of fluid spray and drying medium.
- c. Recovery of the dried product.

2.3.1.1 DISPERSION

The fluid may be dispersed (sprayed) by pressure nozzles or by centrifugal disc atomizers. In pressure nozzles the dispersion is obtained by forcing the fluid through a very small orifice at a high velocity. Centrifugal disc atomizers disperse liquids by causing them to become extended on a rapidly rotating disc into thin sheets which are discharged into the surrounding hot gases at high speeds from the periphery of the disc.

2.3.1.2 CONTACT

Contact between fluid spray and drying medium is usually accomplished in a chamber, the size and shape of which depends upon the spray shape, particle velocity, and drying time. The hot gas drying medium (up to 1400°F) may be the product of combustion from oil, gas, or coal-fired furnaces. The maximum usable temperature is dependent on the thermal stability of the material being dried.

2.3.1.3 RECOVERY

The dried product is customarily recovered as a powder continuously from the bottom of the spray chamber. If the dried material is carried along with the exit gases it may be recovered in a "cyclone" (centrifugal) solids separator.

Spray-drying has been successfully applied on an industrial scale to the drying of such items as inorganic salt solutions, organic compounds having temperature sensitivity, eggs, skim milk, detergents (powders), and coffee. Spray drying technology is well developed and widely applied in the chemical process industries.

Operating data reported in the literature were used to establish a preliminary value for the evaporative capacity of a typical unit volume in a spray-drier (Ref. 10):

<u>Material</u>	<u>Dryer Diameter in Feet</u>	<u>Percent Solids in Feed</u>	<u>Evaporative Rate in Pounds per Hour</u>	<u>Inlet Gas Temperature</u>
Inorganic Salt	18	25	7800	900°F

A representative value for the evaporative rate per cubic foot of drier volume may be calculated assuming the drier height equal to the diameter. From our example above, the volume of the 18 foot diameter drier is 4600 cubic feet and the evaporative rate based on the above figures is 1.7 pounds of water per cubic foot of volume.

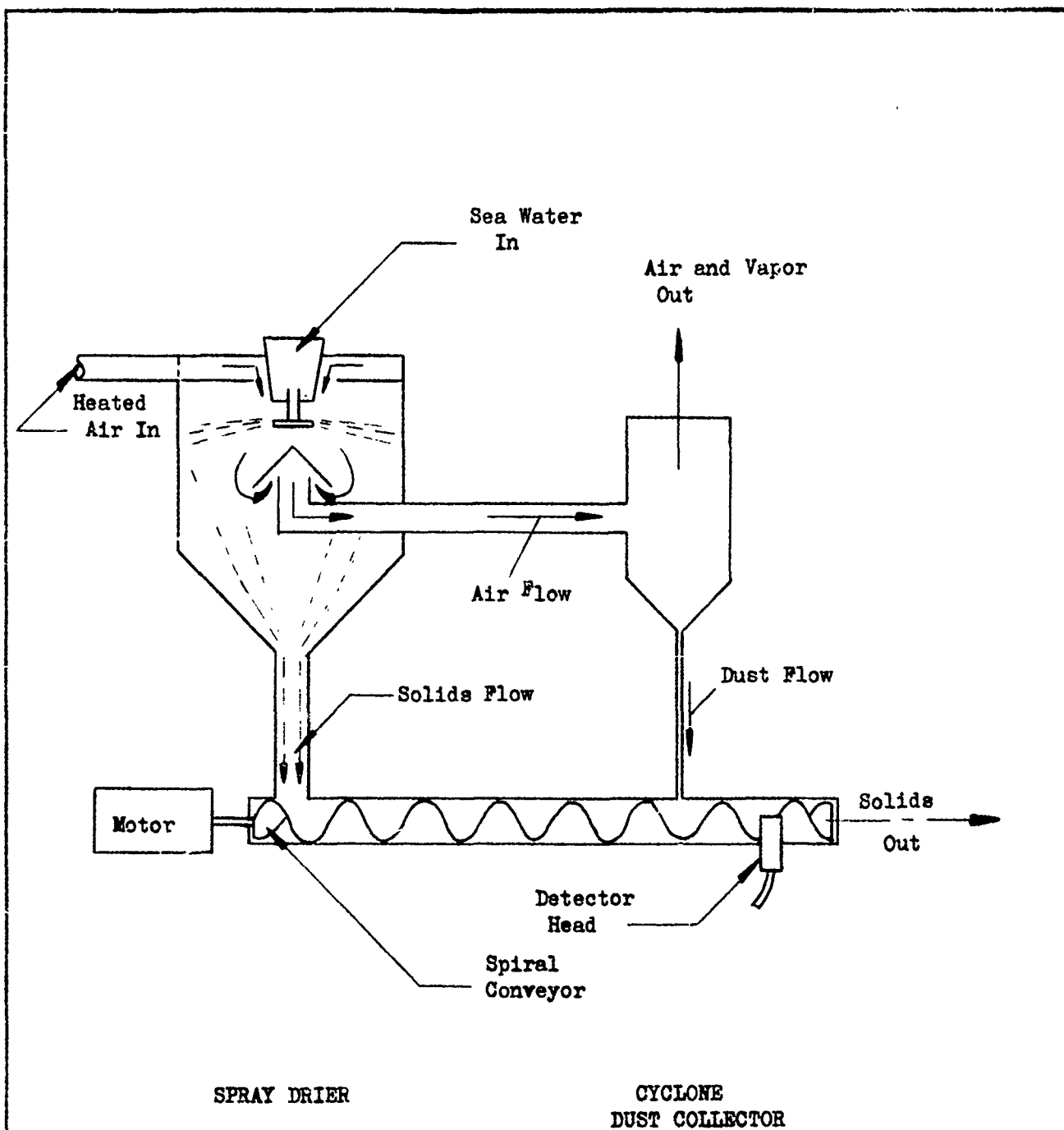
2.3.2 PERFORMANCE CRITERIA

2.3.2.1 SYSTEM CONSIDERATIONS

System requirements for 200 counts in 1, 5, and 10 minutes were not computed because detector efficiencies for the transmission of gamma radiation through dried sea water solids (and detection with a sodium crystal) are not available at this time. This computation is pending availability of the above data.

2.3.2.2 EQUIPMENT CONSIDERATIONS

The evaporation of sea water to dried solids by means of continuous spray-drying techniques is technically feasible. Data exists for the design of these units and there is no question that such a unit could be obtained on a guaranteed performance basis. (See Figure 3).



CAIC		REVISED	DATE	Figure 3. Spray Drier Flow Diagram	D2-7908
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2.3.3 CONCLUSIONS AND RECOMMENDATIONS

Based on the dissolved solids content of 35 grams total salts per kilogram of sea water (solution) (Ref. 2), the weight ratio of solids concentration in dried solids to solids concentration in sea water is 28.5 to 1 while the volume ratio of solids concentration in sea water is 59 to 1. Therefore, the maximum concentration ratio that it is possible to obtain by any method in which the sea water solids are dried and recovered is 28.5 to 1 by weight and 59 to 1 by volume.

It is recommended that data for efficiency of detection of gamma radiation as a function of sample size for dried activated sea water solids be obtained experimentally in order for system analysis to be completed.

3. ION-EXCHANGE TECHNIQUES

3.1 BACKGROUND (Ref. 11)

The mechanism of ion-exchange in ion-exchange resins is analogous to the exchange of crystal lattice ions. The ion-exchange resins may for practical purposes be considered high molecular weight polymeric electrolytes. It is commonly accepted that the exchange of ions with these resins takes place throughout the whole general structure of the resin and is not limited merely to the surface of the material.

3.1.1 STRUCTURE OF ION-EXCHANGE RESINS (Ref. 12)

An ion-exchange resin can be described simply as a high molecular weight polymer containing ionic groupings as integral parts of the

polymer structure. The polymeric structure is so highly cross-linked that solubility of the materials is negligible.

The actual physical form of this material can be varied to produce such physical configurations as membranes and granules. Recently some work has been done to incorporate ion-exchange groups on cellulose in order to produce an ion-exchange fabric. Thus in a broad sense, ion-exchange resins are high polymers which contain certain reactive groups which give to the material the unique "ion-exchange" capability.

Since the "ion-exchange" capability is not limited to the surface of this resin but theoretically exists throughout the resin mass, it is apparent that the degree of cross-linking of the high-polymer will have an effect on the rate of ion-exchange as limited by mass-diffusion rates through the resin. Decreasing the particle size of a resin will therefore result in less time being required for the material to reach equilibrium with a solution.

3.1.2 KINETICS OF ION-EXCHANGE

The over-all exchange process may be divided into five distinct steps:

- a. Diffusion of ions through the solution to the surface of the exchange particles.
- b. Diffusion of the ions through the particle.
- c. Exchange of these ions with those already on the exchanger.
- d. Diffusion of the displaced ions through the exchanger.
- e. Diffusion of these latter ions through the solution.

It is generally accepted that the over-all rate of the ion-exchange reaction is determined by diffusion rate rather than by the actual

exchange reaction at the site (Refs. 13, 14). Some effort has been expended in the past in attempting to establish concentration levels at which the different mechanisms might be expected to be controlling. It has been established that (in general) at concentrations below 0.003 molar the diffusion of ions through the solution film about each particle is considered to be the rate determining process, while at concentrations above 0.1 molar, the diffusion of ions through the particle becomes the rate-determining process. The fact that both diffusional processes are rate-determining between 0.007 molar and 0.1 molar is of importance since most ion-exchange applications fall within this range.

It is reported that at normal degrees of cross-linking for commercial resins the self-diffusion constants (diffusion of an ion within the particle) are about one-tenth to one-fifth the constant of diffusion in free solution. However, by decreasing the cross-linking of the resin, the rate of diffusion may be made to approach that in free solution.

Summarizing for kinetics of ion-exchange, the rate of diffusion of an ion within an ion-exchange resin particle is dependent upon:

- a. Degree of cross-linking of the resin.
- b. Ionic charge of the diffusing ion.
- c. Exchange capacity.
- d. Ionic compositions of the resin.

The fact that, in general, rates of exchange in ion-exchange resins increase for most conditions with decreasing particle size and increasing temperature is strong evidence for a particle-diffusion controlled process.

3.1.3 SELECTIVITY (Ref. 15)

Since the selectivity of a resin for one ion over another is largely a function of the size of the ion, as the cross-linkage decreases and the structures become more porous, selectivity decreases. The degree of cross-linking also has an effect on the purely physical properties of this resin. Resins with a very low degree of cross-linking are practically gels, becoming swollen and soft in water. Highly cross-linked resins are somewhat brittle.

3.2 PERFORMANCE CRITERIA

3.2.1 INTRODUCTION

This section will consider the following operational modes for the use of ion-exchange techniques in the concentration and recovery of sodium-24 and chlorine-38 activity from sea water:

- a. Concentrate activity on resin and detect on resin.
- b. Concentrate activity in the regenerating solution and detect in solution.
- c. Concentrate activity in the regenerating solution, evaporate to dryness and detect in the residue.
- d. Remove the potassium-40 background and detect in sea water without concentration.

3.2.2 CONCENTRATE ACTIVITY ON RESIN AND DETECT ON RESIN (Ref. 16, 17, 18)

Ion-exchange operations in general have been most successful when used for the treatment of solutions of relatively low concentration--generally on the order of a few parts per thousand. On this basis sea water is an extremely concentrated solution containing approximately 35 parts per thousand total dissolved salts (Ref. 19). By simple computation it has been shown that the volume concentration

ratio which it is possible to obtain by this technique for recovery of chlorine-36 using a modern high capacity ion-exchange resin is approximately two. Results of this computation are summarized in Table 2 (Ref. 16).

It is evident from a consideration of the computed activity volume ratio (final column in Table 2) that there is little or no advantage to be gained by concentrating the chlorine-38 activity for solid phase detection on a resin. Therefore, this method will not be considered further. Approximately the same concentration ratio applies to sodium-24 concentration by this technique.

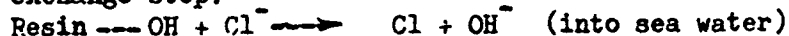
3.2.3 CONCENTRATE ACTIVITY IN THE REGENERATING SOLUTION AND DETECT (Refs. 16, 17, 18, 19)

Another means of concentrating the activity content of irradiated sea water by ion-exchange is to carry out the exchange step (thus placing the desired ion on the resin) and then to regenerate the resin. The net result is that the desired ion is concentrated in the regenerating solution. This scheme simply carries out the general over-all ion-exchange-regeneration cycle wherein the ion is removed from the solution by the resin and then eluted from the resin--the net result being:

- a. The ion originally in the sea water is now concentrated in the regenerant effluent.
- b. The exchange resin has been regenerated to its initial state and is once again capable of taking the desired ions from solution.

Stated in equations: (for chloride-ion exchange)

Ion-exchange step:



Regeneration step:



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TABLE 2. Concentration of Chlorine-38 on High Capacity Ion-Exchange Resin
(Dowex I).

<u>Activity Microcuries per Milliliter</u>	<u>Volume of Sea Water Sample (Liters per Cu. Ft.)</u>	<u>Resin Volume Corresponding to Sea Water Sample</u>	<u>Activity Volume Concentration Ratio: Resin to Sea Water</u>
10^{-9}	500/15.9	7.35	2.16
10^{-8}	50/1.59	0.73	2.16
10^{-7}	5/0.159	0.073	2.16

Calculations indicate that the volume concentration ratio which it is possible to obtain for chlorine-38 by this means is approximately five. Approximately the same concentration ratio applies to sodium-24 concentration by this technique. Results of these computations are given in Table 3.

As indicated in Table 3, activity volume concentration ratio is approximately five. This would permit some gain in detector efficiency.

3.2.4 CONCENTRATE ACTIVITY IN THE REGENERATING SOLUTION, EVAPORATE TO DRYNESS AND DETECT

From work of the proceeding section, the volume concentration activity ratio which it is possible to obtain by eluting the exchange resin with 10 percent (weight) of sodium hydroxide is approximately five. The material balance for the regeneration step would appear as shown in Figure 4.

The ratio of chloride ion (after/before exchange) is obtained by dividing the concentration of chloride ion in the stream out of the ion-exchange column (Figure 4) by the concentration of chloride ion in sea water. This results in a chloride ion concentration ratio (after/before exchange) of approximately five on a weight basis and 4.4 on a volume basis.

Evaporating the solution from the ion-exchange column to dryness will result in an additional gain in concentration ratio. This is the recommended next step. Computations are presented here:

TABLE 3. Summary of Results, Concentration of Chlorine-38 Activity onto Ion-Exchange Regenerating Solution.

<u>Activity Microcuries per Milliliter</u>	<u>Volume of Sea Water Sample in Liters per Cu. Ft.</u>	<u>Volume of Regenerating Solution in Cu. Ft. (10% NaOH)</u>	<u>Activity Volume Concentration Ratio</u>
10^{-9}	500/15.9	3.6	4.4
10^{-8}	50/1.59	0.36	4.4
10^{-7}	5/0.159	0.036	4.4

Basis: 10 weight percent sodium hydroxide solution input for regeneration.

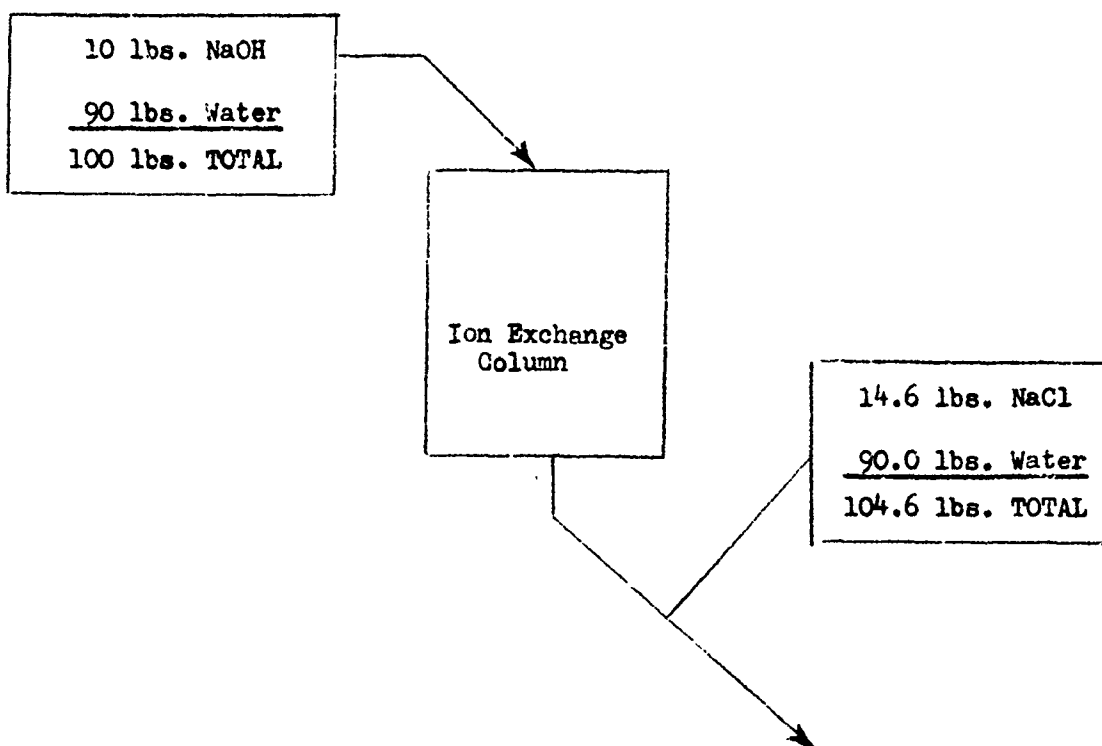


FIGURE 4. Material Balance for Regeneration.

$$1) \text{ Concentration of chloride ion in sea water} = \frac{19.37 \text{ grams chloride ion}}{1000 \text{ grams sea water}}$$

2) Equivalent concentration of sodium chloride in sea water (based on chloride ion concentration)

$$= \frac{19.37 \text{ grams chloride ion}}{1000 \text{ grams sea water}} \times \frac{58.5 \text{ grams NaCl}}{35.5 \text{ grams chloride ion}}$$

$$= \frac{32 \text{ grams NaCl}}{1000 \text{ grams sea water}}$$

$$3) \text{ Weight ratio concentration of NaCl in solid NaCl to NaCl in sea water} = \frac{1000}{32} = 31.2$$

4) Volume ratio concentration of NaCl is computed as follows:

$$\frac{19.37 \text{ grams chloride ion}}{976 \text{ milliliters total sea water volume per kilogram}}$$

$$= \frac{0.01985 \text{ grams chloride ion}}{\text{milliliter sea water}} = \frac{0.0327 \text{ grams NaCl (equiv.)}}{\text{milliliter sea water}}$$

Converting to volume concentration basis:

$$\frac{0.0327 \text{ grams NaCl (equivalent)}}{\text{milliliter sea water}} \times \frac{1 \text{ milliliter NaCl}}{2.165 \text{ grams NaCl}}$$

$$= \frac{0.0151 \text{ milliliters NaCl (equivalent)}}{\text{milliliter sea water}}$$

5) Volume concentration ratio of chloride ion attainable by use of ion-exchange column and evaporation to dryness:

$$\frac{\frac{1000 \text{ milliliters NaCl}}{1000 \text{ milliliters total volume}}}{\frac{0.0151 \text{ milliliters NaCl}}{1 \text{ milliliter sea water}}} = 66$$

The evaporative step can be carried out using spray-drying techniques.

The computed data for this scheme are listed in Table 4. The volume concentration activity ratio attainable is approximately 66, a significant increase in concentration.

TABLE 4. Estimate of Requirements to Concentrate Chlorine-38 as Sodium Chloride Using Combined Ion-Exchange and Spray Techniques.

Activity in Micro- curies per Mil- liliter	Sample Volume in Liters	Weight of Resin per Sample (Pounds)	Weight of Solid NaOH to Elute Chloride Ion (lbs)	Pounds of 10% NaOH Solution Required	Pounds of Water to be Eva- porated	BTU per Sample at 1250 lb. of Sample	Pounds of No. 2 Fuel Oil Required	Ratio of Chloride Ion Con- centration Weight to Volume
10^{-9}	500	327	24.65	246	222	2.7×10^5	15	31.2/66
10^{-8}	50	32.7	2.46	24.6	22.1	2.7×10^4	1.5	31.2/66
10^{-7}	5	3.27	0.25	2.46	2.21	2.7×10^3	0.15	31.2/66

3.2.5 REMOVAL OF THE POTASSIUM BACKGROUND AND DETECT ON RAW SEA WATER WITHOUT CONCENTRATION

This method is to be evaluated based on the use of two resins:

- a. A commercial high-capacity ion-exchange resin (Dowex 50) (Ref. 15).
- b. A potassium-specific resin reported in the literature (Ref. 20).

In conventional ion-exchange, the natural separation factor of the resin is used to exchange the potassium ion (and some sodium ion) from the sea water by simply passing the fluid through the resin. Since sea water is presumed to be of constant composition, the separation factor obtained will be a function of the resin properties and operating parameters.

3.2.5.1 SEPARATION WITH CATION-EXCHANGE RESIN (Ref. 21)

Based on equilibrium data as reported for the system potassium chloride-sodium chloride-water, a preliminary evaluation of the scheme for the removal of potassium from sea water using Dowex 50 resin was carried out. Basic data for sea water are as follows:

<u>Cation--Grams per Liter</u>		<u>Cation Mol-Fraction</u>	
Sodium	11.1	Sodium	0.87
Potassium	0.4	Potassium	0.02
Calcium	0.4	Calcium	0.02
Magnesium	1.3	Magnesium	0.09
TOTAL	13.2		1.00

From these values the material balance shown in Figure 5 indicates the degree of removal of sodium ion when all of the potassium is removed at equilibrium condition.

Basis: 100 lb. Mols of Cations (positively charged ions) dissolved in Sea Water

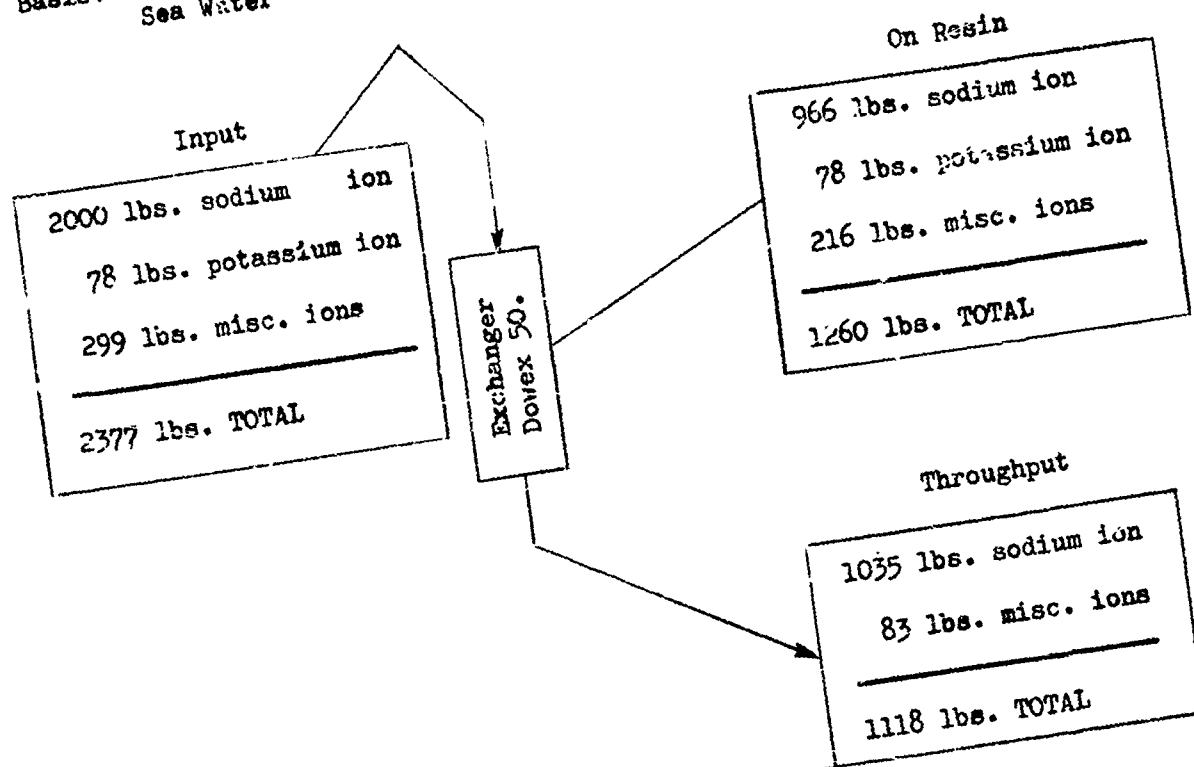


FIGURE 5. An Indication of the Degree of Removal of Sodium Ion when all of the Potassium Ion is Removed at Equilibrium Conditions Using Dowex 50 Resin.

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Based on the equilibrium data reported in the literature, therefore, in removing all of the potassium from sea water using commercial cation-exchange resin, approximately 50 percent of the sodium will be removed from the solution--the remaining 50 percent will be available for detection. (U)

3.2.5.2 SEPARATION WITH POTASSIUM SPECIFIC RESIN (Ref. 20)

Based on data reported in the literature, the material balance shown in Figure 6 indicates the degree of removal of sodium ion when all of the potassium ion is removed at equilibrium conditions using a potassium resin, about 87 percent of the sodium will pass through the resin when all of the potassium is removed. This resin is not commercially available. (U)

3 CONCLUSIONS AND RECOMMENDATIONS

Of the four schemes presented for ion-exchange recovery of activity in sea water, two appear to have potential and should be investigated further: (U)

- a. Concentrate activity into regenerating solution, evaporate to dryness and detect. (U)
- b. Remove the potassium-40 and detect on raw sea water. (U)

It is recommended that experimental work be carried out on the above two methods in order to better evaluate their potential as reinforcing techniques for nuclear submarine detection. (C)

4. PRECIPITATION TECHNIQUES

4.1 BACKGROUND

The separation of sodium and chlorine from sea water by precipitation is technically feasible. The greatest potential lies in the (U)

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Basis: 100 lb. mols of cations dissolved in seawater

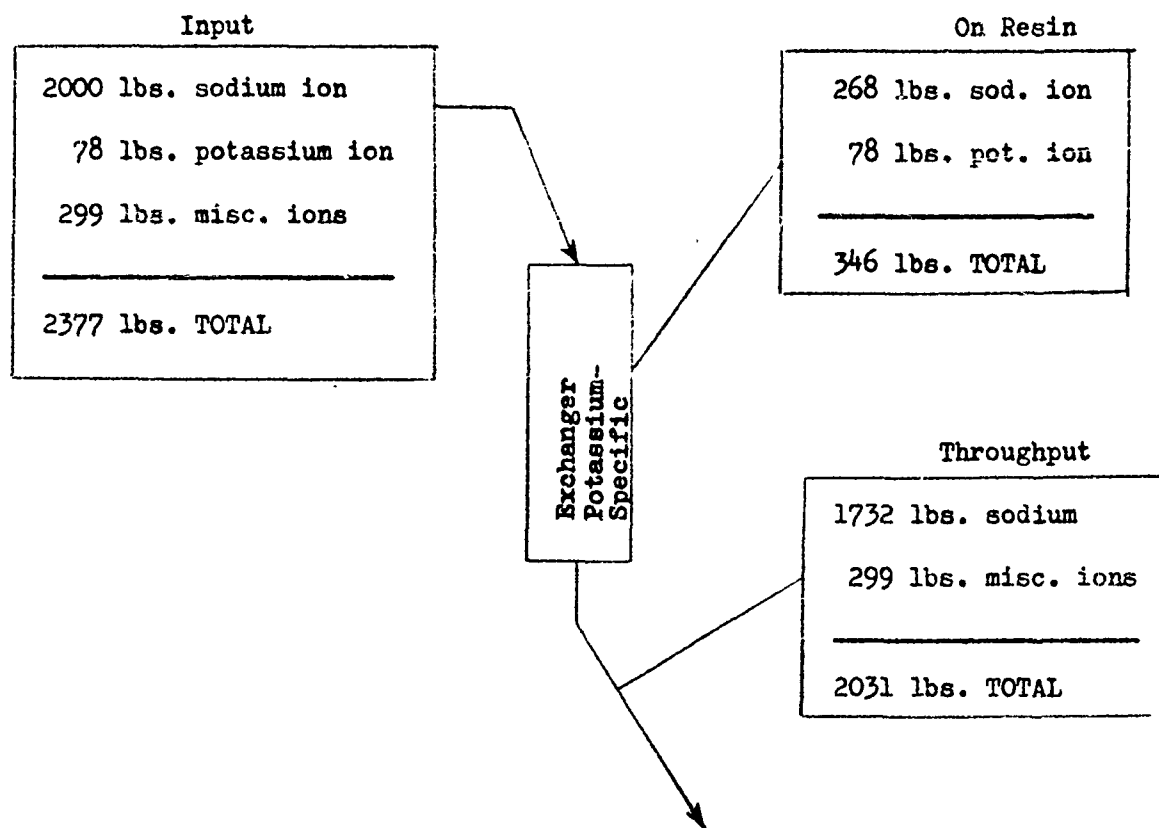


FIGURE 6. An Indication of the Degree of Removal of Sodium Ion when all of the Potassium Ion is Removed at Equilibrium Conditions Using a Potassium-Specific Resin.

precipitation of the sodium as sodium chloride in a concentrated hydrochloric acid solution and in the precipitation of the chlorine as silver chloride, lead chloride, or mercurous chloride in normal sea water.

4.2 CHEMICAL CONSIDERATIONS

Consideration will be given to precipitation of sodium-24 as sodium chloride in a concentrated hydrochloric acid solution and precipitation of chlorine-38 as silver chloride, lead chloride, or mercurous chloride.

4.2.1 PRECIPITATION OF SODIUM-24

From the standpoint of reported solution equilibria, it would appear that the potassium ion will not precipitate from solution under the system conditions necessary for sodium chloride precipitation in the system sodium chloride-potassium chloride-hydrochloric acid. Therefore, a "clean" separation of sodium from potassium should result. This would eliminate a major part of the background problem. There are no "chemical problems" hidden here.

4.2.2 PRECIPITATION OF CHLORINE-38

This is a direct chemical precipitation in sea water and should present no problem insofar as the chemistry is concerned. The precipitate would be filtered, dried, and counted, presumably by automated techniques.

4.3 SYSTEM CONSIDERATIONS

The precipitation of a 95 percent fraction of sodium as sodium chloride from sea water will necessitate acidifying the entire sea

water sample to about 36 percent (by weight) hydrochloric acid using hydrogen chloride gas. The conversion of sea water to 36 percent (weight) hydrochloric acid will require approximately 56 pounds of hydrogen-chloride gas per 100 pounds of sea water. Since hydrogen chloride gas is normally stored at 600 pounds per square inch in steel cylinders at a normal loading of 300 pounds of hydrogen chloride per cubic foot of cylinder volume, approximately two cubic feet of cylinder storage per 100 pounds of sea water sample will be required.

Preliminary conservative computations for required sea water sample size at an induced activity level of 10^{-9} microcuries per milliliter indicates that 120 pounds of hydrogen chloride would be required for a single sample capable of generating 200 counts per minute.

The recovery of the hydrogen chloride from the spent sea water sample is a technical possibility through the use of advanced distillation methods. It has been concluded, however, that the complexity and power requirements for such a proposal reduce the system practicality to well below that of other available methods.

The precipitation of chlorine-38 as silver, lead, or mercurous chloride will require that suitable reagents be available. These will probably be the nitrate salts. From a purely mechanical standpoint, the in-process time for chlorine precipitation could probably be reduced to the order of a minute or less.

For induced activity levels of 10^{-9} microcuries per milliliter, preliminary conservative computations indicate that approximately 90 pounds of precipitating reagent (silver nitrate for example) will be

required for a sample capable of generating 200 counts per minute. The regeneration of the reagent for reuse is considered to not have a high order of practicality for this system.

4.4 CONCLUSIONS

The concentration of the activity in sea water by the chemical precipitation of the sodium and chloride ions using the chemical systems described above appears to have a lower level of practicality than other methods considered to date.

5. FREEZING TECHNIQUES

5.1 BACKGROUND (Ref. 22)

The reason for evaluating a freezing method for the concentration and recovery of solids dissolved in sea water is that the quantity of energy to be removed in the freezing of a given quantity of water is only a fractional part of the energy which is required to evaporate a similar quantity of water. For example, at one atmosphere total pressure, the latent heat of fusion for water is approximately 150 BTU per pound of water, compared to nearly 1100 BTU per pound for the latent heat of vaporization--a factor of over seven to one. Such a difference in the basic thermodynamic requirements justifies giving some serious consideration to methods for concentrating the solids dissolved in sea water by freezing techniques.

5.2 CURRENT STATUS

Work has been carried out on the problem of obtaining potable water from sea water through freezing techniques (Ref. 22). Most of these studies deal with equipment evaluation from which a considerable quantity of data has been developed.

There are at the present time three major problems which must be solved before any mechanically feasible process can be worked out-- these are:

- a. Separation of pure ice crystals from a brine solution.
- b. Heat transfer improvement with regard to magnitude and nature of the job.
- c. Developing a continuous process.

5.3 CONCLUSIONS

The concentration of the activity in sea water by freezing techniques is in an early stage of development and cannot be considered practical at this time.

6. LIQUID-LIQUID EXTRACTION TECHNIQUES

6.1 BACKGROUND (Refs. 23 - 25)

Liquid-Liquid extraction is the term applied to an operation wherein a material dissolved in one liquid phase is transferred to a second phase. For the purpose of concentrating and recovering the activity in sea water, this would entail either: (1) The removal of the dissolved solids from sea water, or (2) The removal of the sea water from the dissolved solids.

Liquid-Liquid extraction consists of three basic steps:

- a. Mixing or contact of the solvent with the solution to be treated so as to transfer the solute from solution to solvent.
- b. Separation of the liquid solution phase from the liquid solvent phase.
- c. Recovery of the solute from the solvent.

Solvent extraction is a recognized unit operation and is a technique widely used in the chemical industry.

6.2 CURRENT STATUS

As indicated above, there are two approaches to the solvent extraction method for desalination:

- a. That of removal of salt from water.
- b. Removal of the water from salt.

The first method is at present not feasible since a liquid solvent which would extract salt from water and from which the salt could then be in some manner removed during the regeneration step is not known. The second method--extracting water from the salt--shows more promise at this time. The solvent required must have certain general properties listed here:

- a. The solvent must selectively dissolve appreciable quantities of water from a saline solution.
- b. The effect of temperature on the solubility must be such that most of the dissolved water will form a separate phase with reasonable temperature change.

A large number of organic compounds have been evaluated with regard to these properties. The most promise appears to be in compounds containing a basic nitrogen atom (amines), and in several types of ethers. To date nothing of practical interest has been reported.

6.3 CONCLUSIONS

At the present time the concentration of solids dissolved in sea water by liquid-liquid extraction appears to be impractical. This type of process will possess considerable potential when an appropriate solvent is discovered or synthesized.

7. MEMBRANE PROCESS (ELECTRODIALYSIS AND OSMOSIS) TECHNIQUES

7.1 BACKGROUND (Refs. 26, 27)

Dialysis is the separation of substances of different molecular sizes in a liquid solution by virtue of their different rates of diffusion through a suitable membrane. Dialysis has found use on a laboratory scale in the removal of electrolytes and the smaller water soluble organic molecules from biological materials such as serums, proteins, hormones, and enzymes. Practical industrial applications are few owing to the low rates of separation which can be obtained and to the relatively poor selectivity of currently available membranes.

7.2 CURRENT STATUS

When electrolytic crystalline substances are to be separated in solution by dialysis methods, the rate of removal of ions can be greatly increased by imposing an electromotive force across the membrane. The ions then move through the membrane towards the electrodes much more rapidly than in ordinary dialysis.

Devices and processes using recently developed ion-exchange membranes have been used for converting brackish and sea water to potable water by converting the saline water feed into a potable demineralized stream and brine. Thus far, significant achievements have been recorded for the production of fresh water from brackish water--3 to 5 parts per thousand of salts, and from sea water--approximately 35 parts per thousand of salts (Ref. 28).

Other membrane processes (osmosis for example) are in a relatively early state of development and show little if any promise for utilization on any industrial scale at this time.

7.3

EVALUATION

The successful utilization of electrodialysis for the production of potable water from brackish or sea water must be analyzed with regard to actual operational data in order to establish the level of potential for possible utilization in the concentration of the activity in sea water.

Data for the production of potable water from brackish water indicate that some degree of concentration is obtained in one of the two exit streams--these data are given in Figure 7 (Ref. 29).

The scale concentrations shown are typical for electrodialyzer performance. Note that the "concentrate" stream has a salt concentration of 14 to 16 parts per thousand for operation on brackish water (3.8 parts per thousand salts). Data for operation on sea water show only a little enrichment of salt content of the brine. This indicates that with present membranes the production of a concentrated brine from sea water by electrodialysis has not been accomplished.

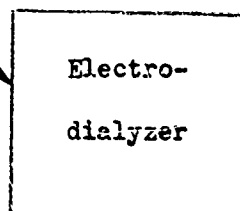
7.4

CONCLUSIONS

Based on operational data for a 24,000 gallon per day pilot plant operating on brackish water (Ref. 29) and on a 172,000 gallon per day plant operating on sea water (Ref. 30) it is concluded that at the present time, the production of a concentrated brine from sea water by a membrane process is not practical.

Brackish Water Input

3.8 ‰ salts



Electro-
dialyzer

1 ‰ Potable
Water Salts
800 gal/min

14-16 ‰
Brine (concentrate)
200 gal/min

FIGURE 7. Concentration Obtained by Electrodialysis.

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NUMBER D2-7908 MODEL NO. _____
TITLE SPECTRAL DATA FOR VOLUME DISTRIBUTED SOURCES, K^{40} , Ce^{38} , AND Na^{24} , WITH SEVERAL SIZES OF SODIUM IODIDE, PLASTIC AND LIQUID SCINTILLATION DETECTORS. JULY 1959-SEPTEMBER 1960.

PREPARED BY E. Papadopoulos
PREPARED BY C. M. Proctor
SUPERVISED BY W. E. Nesbitt
APPROVED BY D. A. Hicks (DATE)

8-23241-5470-54053-0

• CHARGE NUMBER

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SECTION TITLE PAGE

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DISCUSSION

This material is being issued as the currently best information available on scintillator response to volume distributed sources of Na^{24} , Cl^{38} , and K^{40} . The data presented herein represents some extensions in the dimensions and types of scintillating material previously reported (in document D2-7908) and will be discussed in detail in a later publication.

Energy resolution has been improved in the case of Pilot B by employing a different reflector material. The previous reflector employed was aluminum foil loosely coupled to the Pilot B. The new data are for the use of Du Pont 29-915 paint (high reflectance white) surrounded with a 1/16 inch layer of MgO . In addition to the 3 x 3 and 5 x 4, 2 inch x 2 inch and 10 inch by 10 inch Pilot B plastic scintillators were employed. Seven multiplier phototubes with balanced outputs were used with the 10 inch x 10 inch Pilot B scintillator.

In addition to the NaI(Tl) crystal data reported in D2-7908 and the revised and extended data for the Pilot B phosphors reported here, the response of a liquid scintillator* in comparable sizes is also reported and also a 12 inch by 12 inch. The containers were made from stainless steel and have a wall thickness of .040 inches.

* Toluene solvent + 4 gms/liter p-terpheryl + 0.2 gm/liter POPOP (1,4 - bis - 2 - (5-phenyloxazolyl) - benzene)

The containers were provided with reflecting walls by employing the following preparation. After sandblasting, five coats of Tygon TP-61 were applied allowing thorough drying between coats. Two final coats of Tygon TP-61 containing liberal amounts of pulverized quartz were then applied. The viewing windows were made of fused quartz for the smaller containers and ordinary window glass for the 12 inch x 12 inch one. The same photomultiplier arrangement that was employed for the 10 x 10 Pilot B was used for the 12 inch x 12 inch liquid.

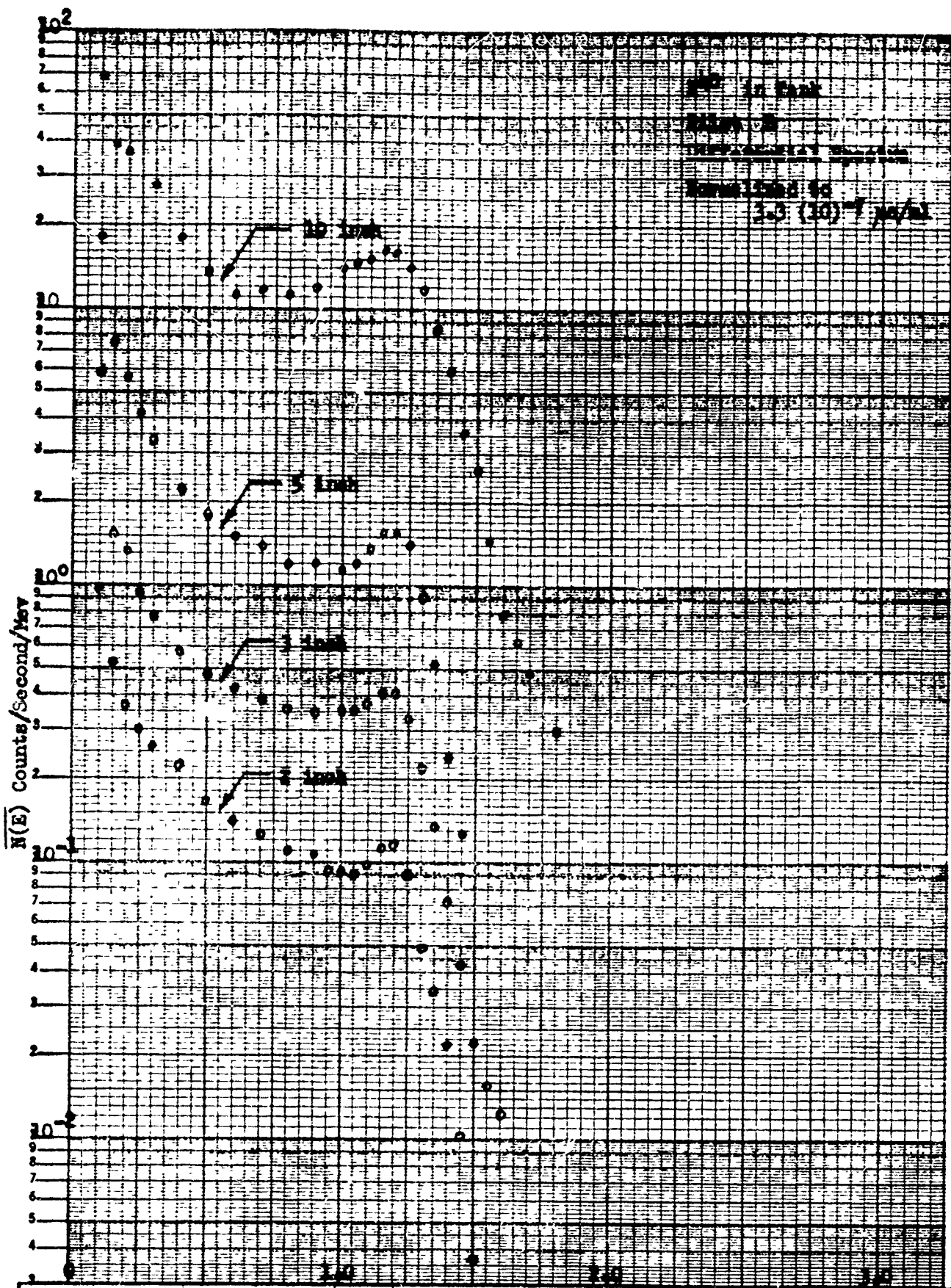
All isotope preparation procedures, mixing, calibration and data preparation are described in D2-7908. In Table 1 the integral count rates above 250 kev are given for three nuclides and a variety of detectors. A rough comparison of the efficiency of the various scintillator sizes and materials can be obtained by using the relative count rates which are given in parenthesis in the table. Comparison of the liquid scintillator and Pilot B data indicates that the liquids offer no advantages over the Pilot B plastic phosphors. On a counting rate per unit volume basis for 5 inch x 4 inch and smaller they seem to be comparable while for the larger sizes (10 inch Pilot B and 12 inch liquid) and the Pilot B seems to be superior. The failure of the larger size liquid to compare favorably with this Pilot B may be due to the poor transmission of the window glass to the scintillator emission spectrum.

TABLE I

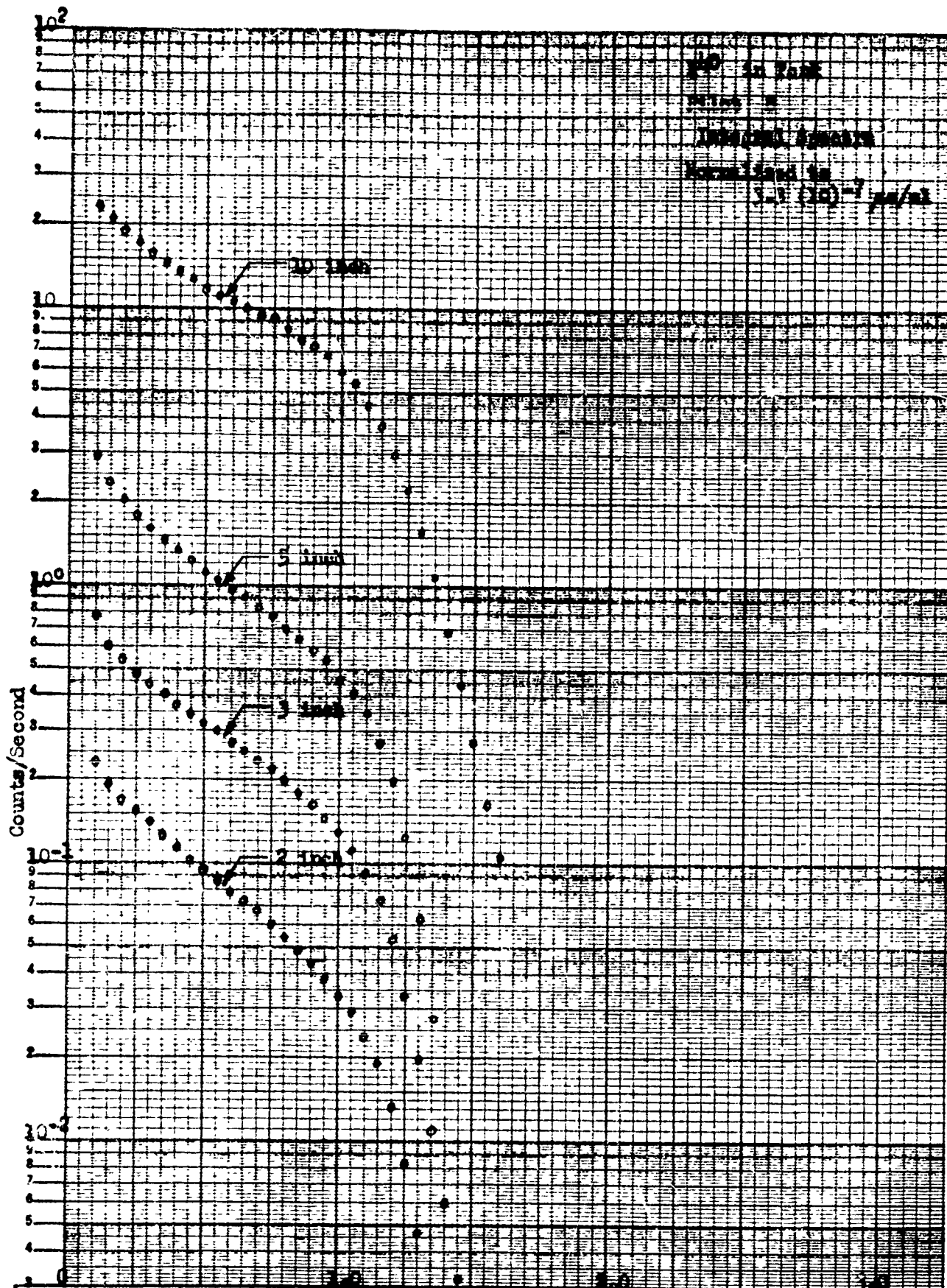
Integral Count Rate above 250 Kev

In counts per second and relative to 5 x 4" NaI(Tl).

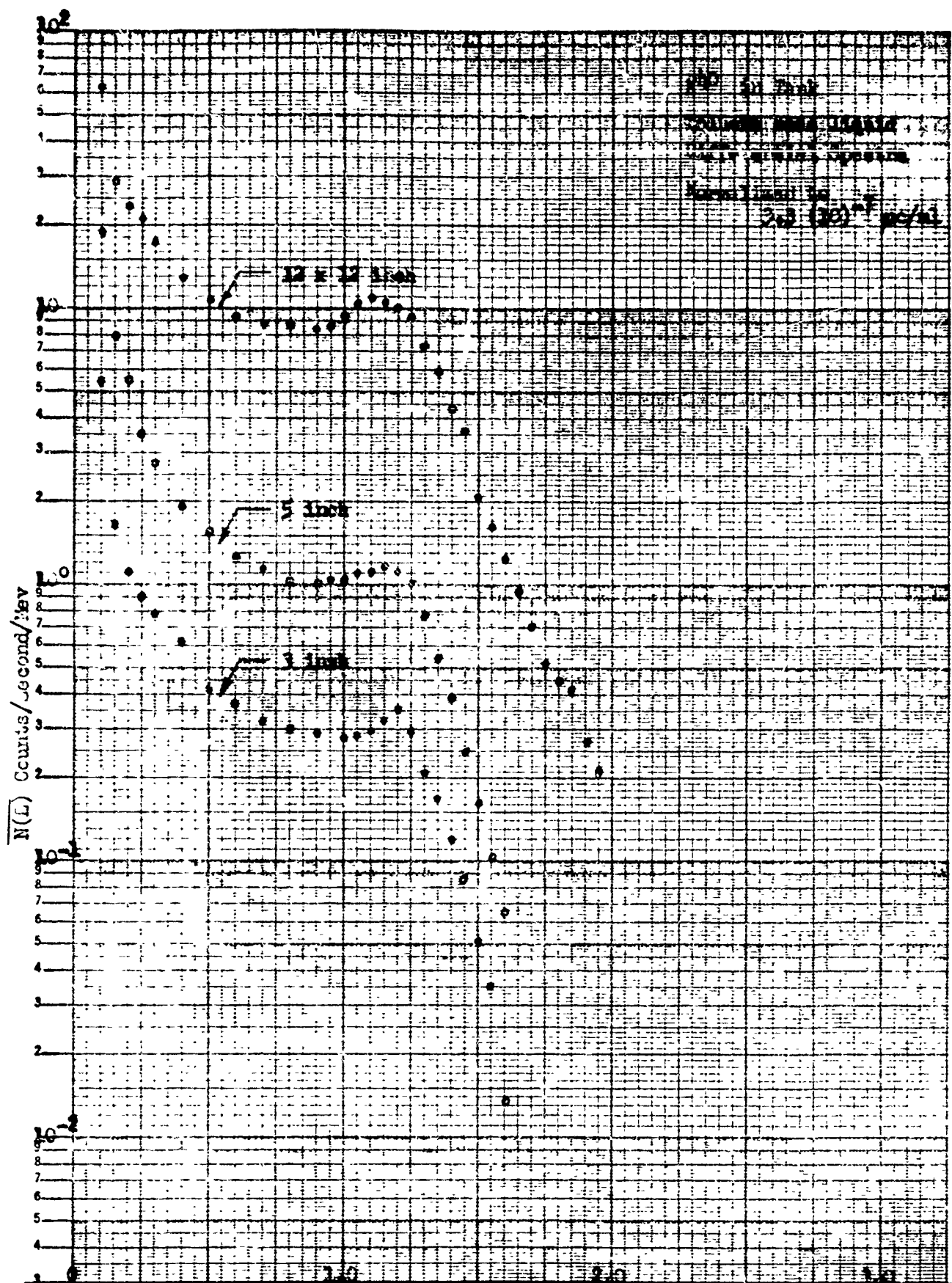
energy Activity	K^{40} (1.46) 3×10^{-7} c/ml		Cl^{38} (1.60, 2.15) 10^{-6} c/ml		Na^{24} (1.37, 2.75) 10^{-6} c/ml	
	C.P.S	Rel	C.P.S.	Rel	C.P.S	Rel
$2 \times 1 \frac{3}{4}$ "						
NaI	0.61	(0.117)	9.2	(0.12)	27.5	(0.11)
Pilot B	0.16	(0.031)	2.25	(0.029)	11.8	(0.047)
Liquid	-	-	-	-	8.5	(0.034)
3×3 "						
NaI	2.15	(0.41)	27	(0.35)	91	(0.36)
Pilot B	0.48	(0.092)	12.3	(0.15)	34.5	(0.14)
Liquid	0.45	(0.087)	-	-	29	(0.12)
5×4						
NaI	5.2	(1.00)	78	(1.00)	250	(1.00)
Pilot B	1.8	(0.35)	39	(0.50)	112	(0.45)
Liquid	1.52	(0.29)	-	-	100	(0.40)
10×10						
Pilot B	17	(3.3)	410	(5.3)	1070	(4.3)
12×12						
Liquid	12.3	(2.4)	-	-	1200	(4.8)



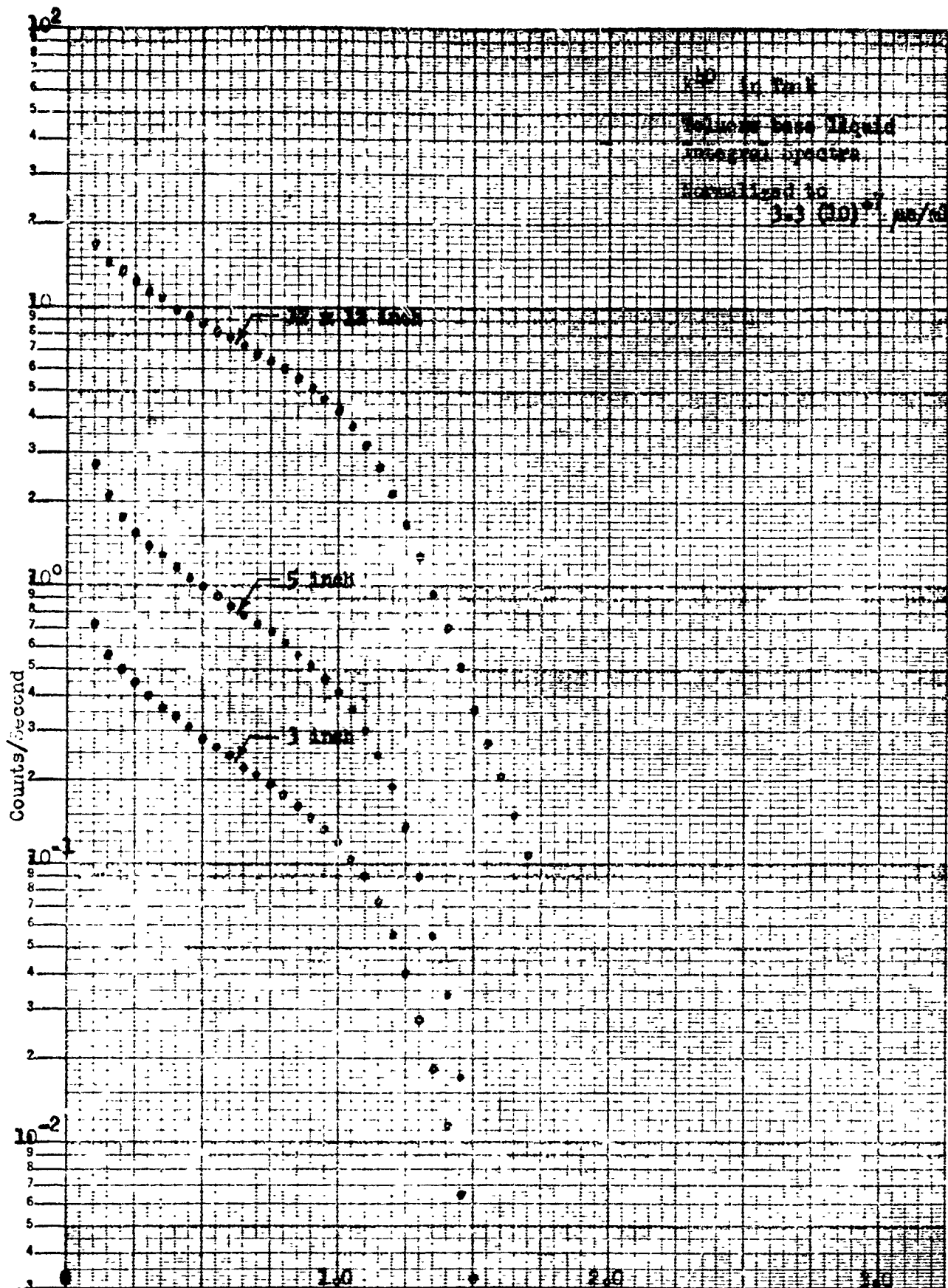
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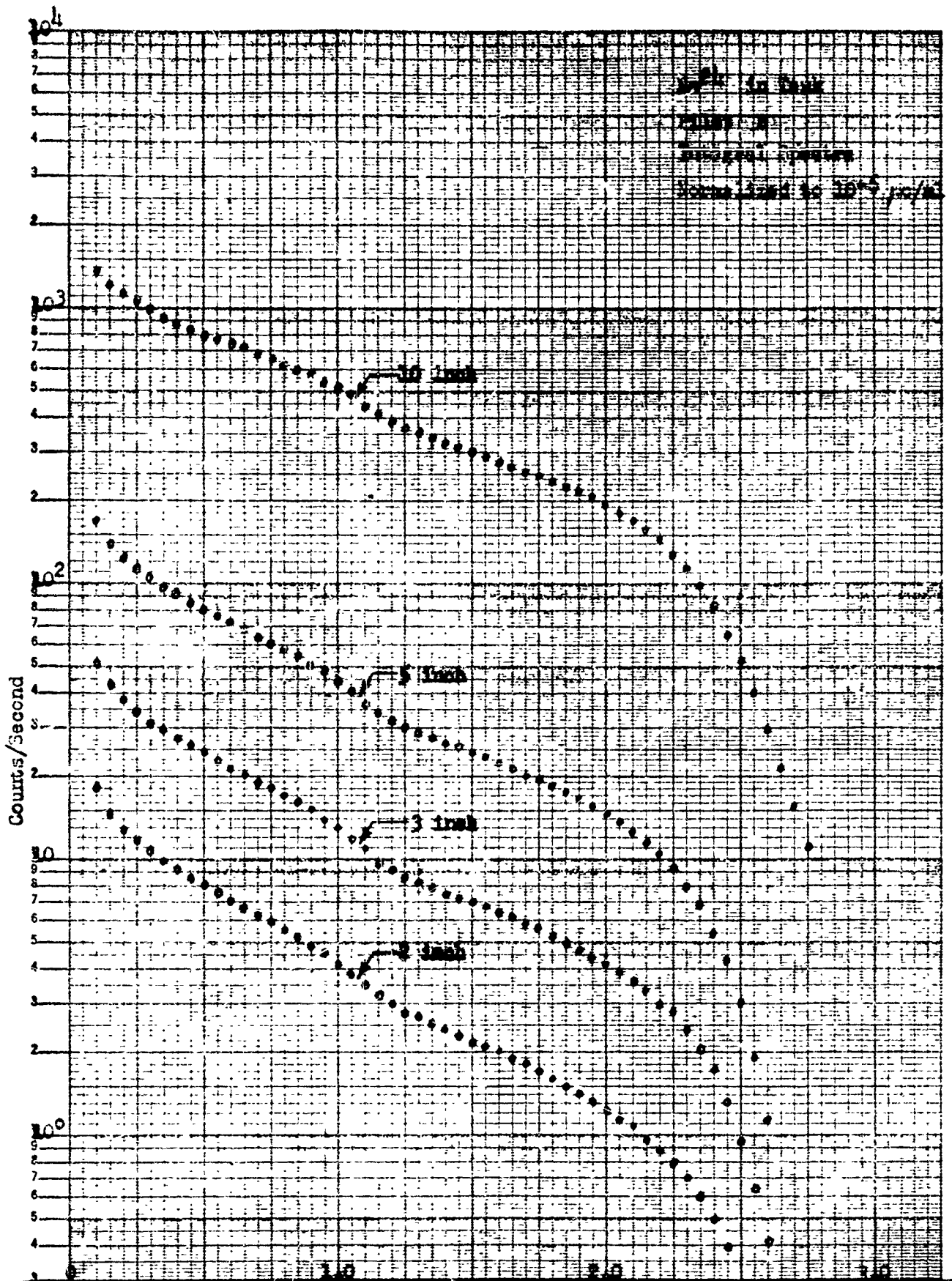
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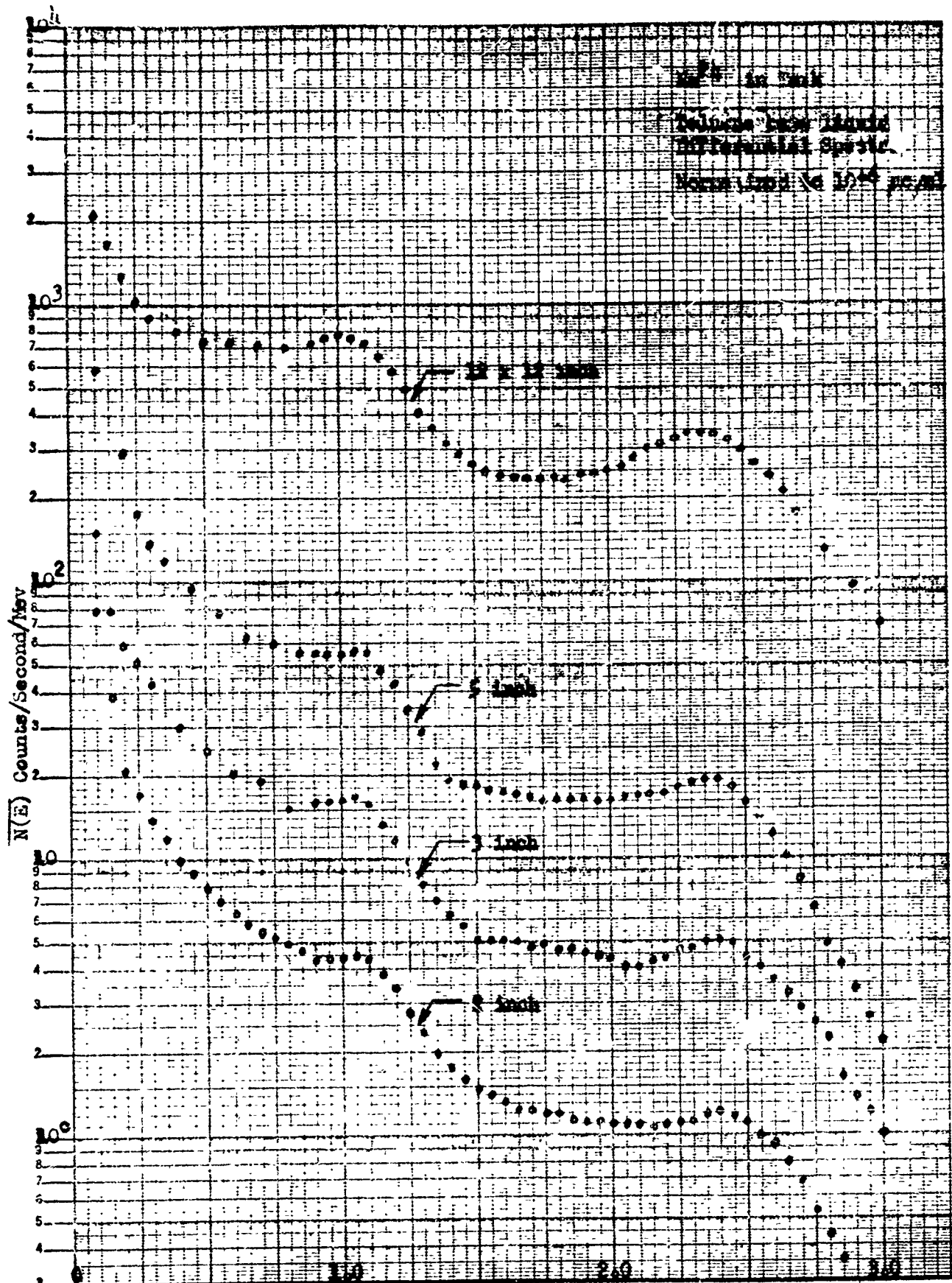
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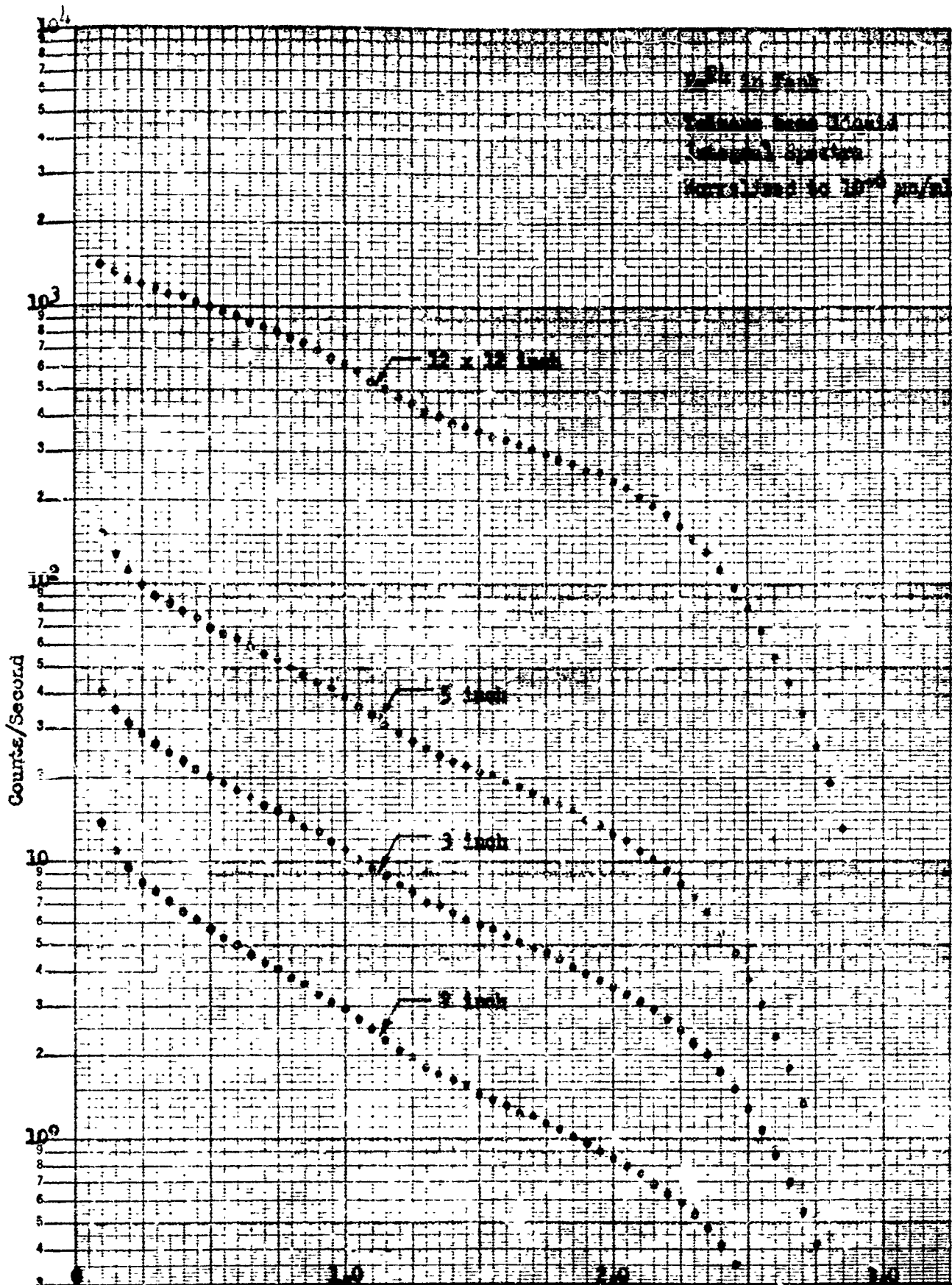
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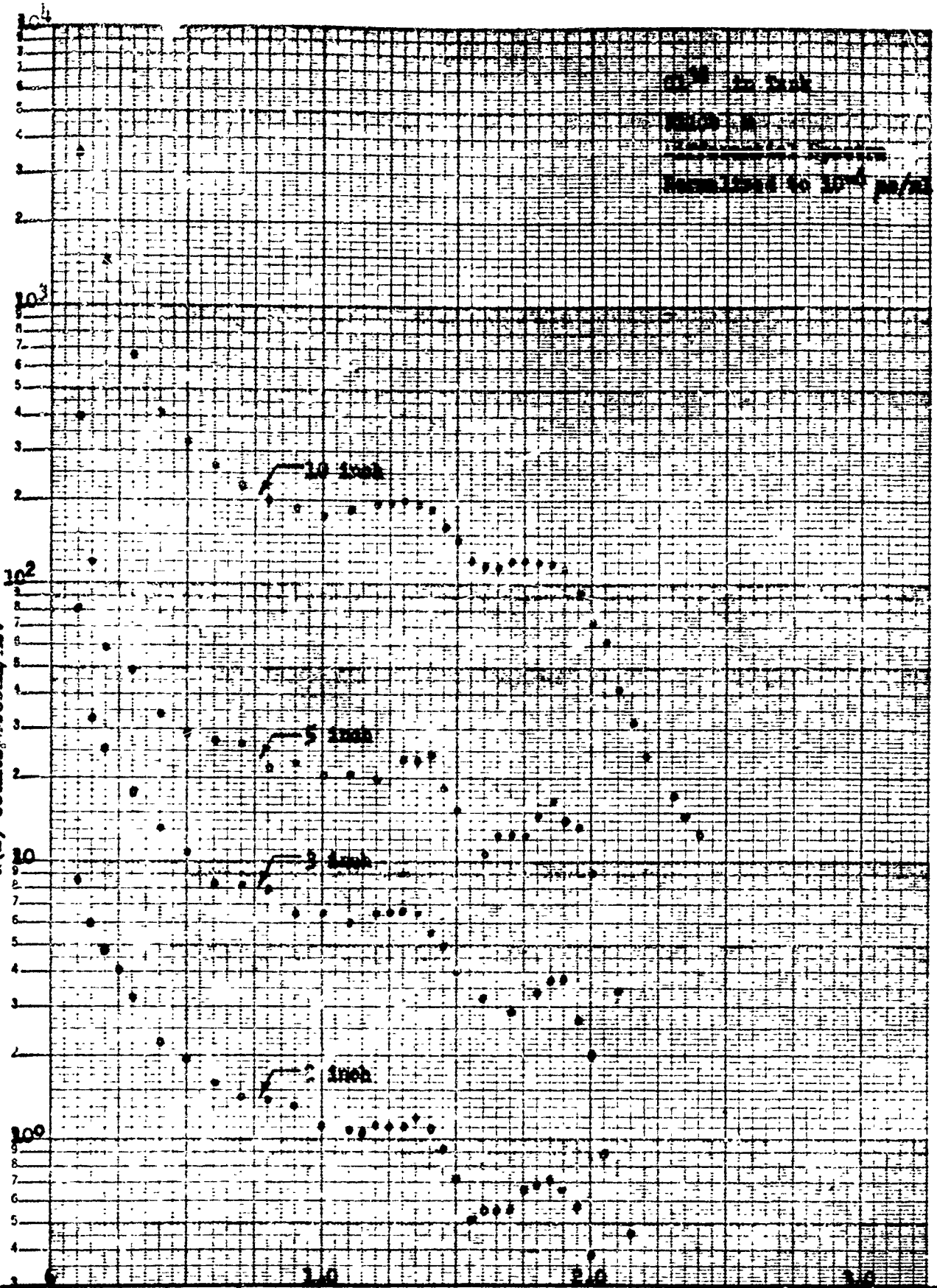


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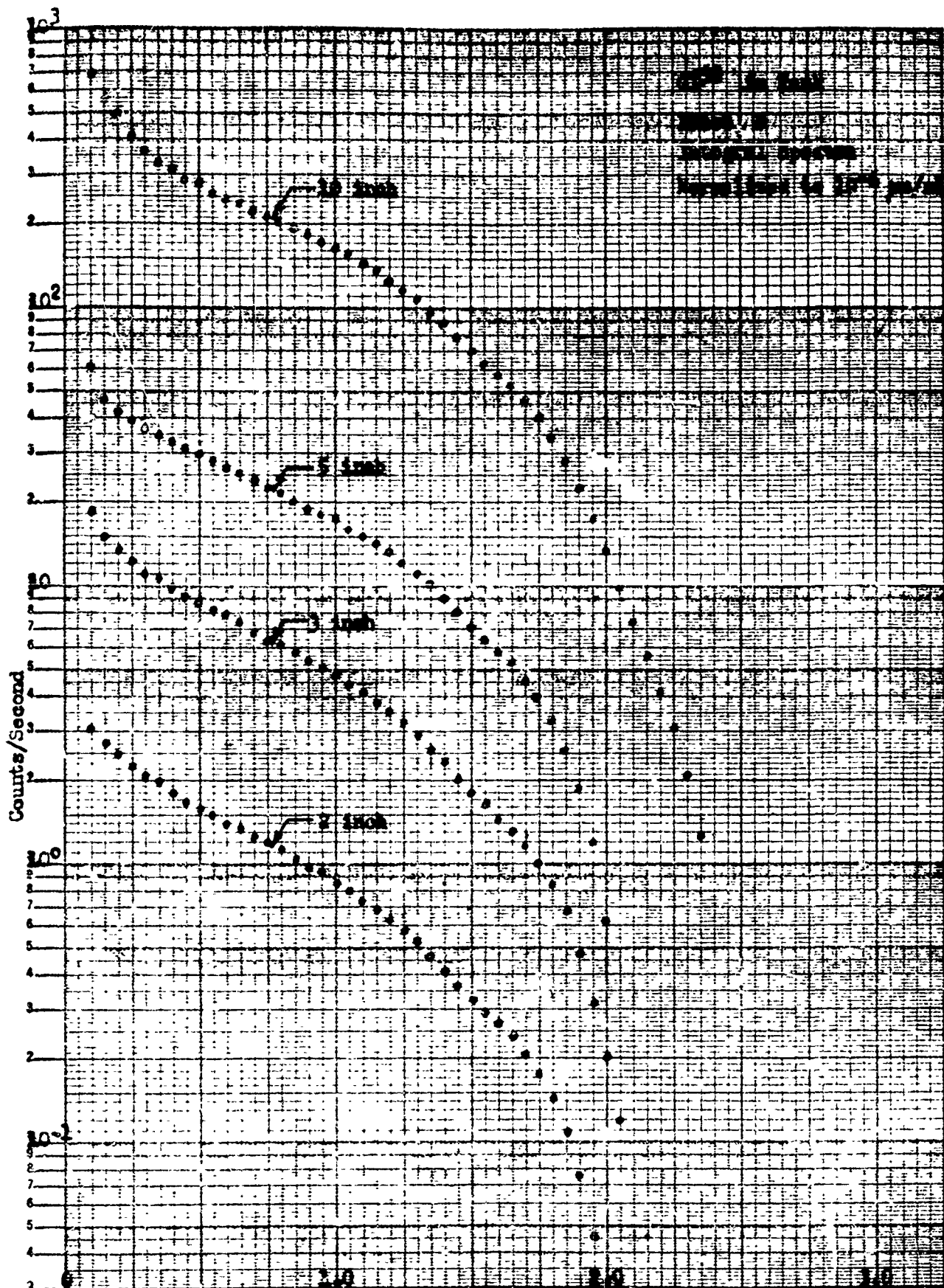


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$N(E)$ Counts/Second/Mev



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